BLACK MAGIC BITUMEN

An archaeometrical approach to 5000 years of bitumen imports in the Persian Gulf

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Proefschrift voorgelegd to het behalen van
de graad van Doctor in de Archeologie

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Nederlandse vertaling:
Zwarte Magie en Bitumen. Een archeometrische blik op 5000 jaar bitumen import in de Perzische Golf

Kaftinformatie: Image courtesy of NASA Visible Earth
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Acknowledgments

This work would not have been possible without the kind help, support, and advice of a great many people. First of all, I would like to express my thanks and appreciation towards my promoter, em. prof. dr. Ernie Haerinck. His teachings motivated me to become active in the archaeology of the Persian Gulf in the first place and to pursue academic research on a doctoral level.

As I became more and more interested in bitumen, I realized I could get the most out of them by approaching them on a molecular and atomic level. This proved to be a great challenge as I am trained in archaeology rather than organic chemistry. But thanks to the kind help from several people from different UGent departments and laboratories, it became possible to conduct qualitative analyses. The research here presented would not have been possible without these people. From the Separation Science Group, I would like to thank prof. Frederic Lynen for allowing me to use the laboratories and facilities and express my special thanks towards Mike de Vrieze and Pieter Surmont for investing both time and effort into this research. From the Isotope Bioscience Laboratory (Isofys) I would like to thank prof. dr. ir. Pascal Boeckx, dr. Samuel Bodé and Katja Van Nieuland for offering me the opportunity to work and analyse at the Isofys laboratory and for their kind help and support. I had a (scientific) blast working at both laboratories thanks to all the kind people over there willing to share their knowledge over a stranded archaeologist!

On the same chemical level, I would like to thank Jacques Connan for his kind help and comments on the many questions and chromatograms I've send him over the last couple of years. His long time experience with analysis on archaeological bitumen proved to be of immense value!

Of course, no analysis without samples. This research was only possible thanks to the numerous samples I received from several excavators who were interested in having their bitumen analysed and put their trust in my skills. So, thanks Philipp Drechsler, Steffen Terp Laursen, Flemming Højlund, Benjamin Porter, Bruno Overlaet, Sabah Jasim, Pierre-Francesco Callieri, Tijs De Schacht & Ernie Haerink!

Over the past years, many friends and colleagues have contributed to this thesis on many different levels: either by engaging in archaeological/chemical/statistical /methodological/whatever discussions and endeavours, by contributing to the fine atmosphere that always lingered in the office, distracting me from work by scientific/cultural-inspired visits to the pub(s), by coaching/participating in the healthy and sportive lifestyle (*mens sana in corpore sano*), or by giving me access to the much-needed shots of rather fine coffee. Naming all of these colleagues and friends would results in a too long list, vastly expending the number of pages of this thesis; therefore a general and sincere THANK YOU to all of you!

Several people have contributed to the reading experience of this work by proof-reading parts of the text and by making valuable comments concerning text and language. So thank you Kim Van Liefferinge, Vince Van Thienen, Thierry Van Neste, Devi Taelman and Helen Taylor. I would also like to thank Gitte Callaert for helping me with the final lay-out of this thesis and making such a nice cover!

Finally, this work would not have been possible without the never-ending support of my girlfriend Karlien Jordens and our son Clement. Admittedly, his support was perhaps carried out unintentionally, but his smile always seemed to lighten up the day. The last couple of months may have been a bit hectic with me more often at the office than at home and with my mind being somewhere in a strange place dominated by Gulf-archaeology, hydrocarbons, isotopes and statistics. But as this thesis is entering its final stages, I'm more than ever looking forward to the impending extension of our family.

Structure of this thesis

Conform with the Ghent University Arts & Philosophy faculty regulations addressing the form and content of PhD's (last updated version of 02-07-2014), this thesis consists of various scientific contributions which are here bundled and presented as one research. All these different essays —printed here as several individual chapters— are the result of one bigger research handling the origin- and character of bitumen on archaeological sites in the Persian Gulf. According to the faculty regulations, at least two of those publications are quoted in either the "Arts and Humanities Citations Index", the "Social Sciences Citation Index", or the "Science Citation Index". The chapters in this thesis derived from these papers are largely unaltered with some exceptions, for instance; the geochemical chapters/papers were stripped of their parts describing analytical methods as these are presented here separately and more extensive in chapter 3.4. The different scientific contributions used in this thesis are the following:

Peer-reviewed Journal articles:

1. Connan J. & Van de Velde T. 2010. An overview of bitumen trade in the Near East from the Neolithic (c.8000 BC) to the early Islamic period. *Arabian Archaeology and Epigraphy*, 21, 1-19. **(A1)**
2. Van de Velde T., De Vrieze M., Surmont P., Bodé S. & Drechsler P. 2015. A geochemical study on the bitumen from Dosariyah (Saudi-Arabia): tracking Neolithic-period bitumen in the Persian Gulf. *Journal of Archaeological Science*, 57, 248-256. **(A1)**
3. Van de Velde T. 2015. Digging into the Ubaid-Period bitumen from Dosariyah. *Proceedings of the Seminar for Arabian Studies.* **(A2, VABB-SHW)**

International Book chapters:

1. Van de Velde T. Accepted for publication. Sourcing the bitumen from Tell F6. *In:* Højlund F. & Abu-Laban A. (eds.) *Tell F6 on Failaka Island. Kuwaiti-Danish Excavations 2008-2012*. Moesgaard: Jutland Archaeological Society.

2. Van de Velde T. & Bodé S. Accepted for publication. Appendix 2: Analysis of Bitumen from the Royal Mounds. *In:* Laursen S. T. (ed.) *The Rise of Kingship and the Early Dilmun State in Bahrain*. Moesgaard: Jutland Archaeological Society.
3. Van de Velde T. & Connan J. Accepted for publication. Bitumen in the 3rd millennium of the Near East. *In:* De Miroschedji P. & Lebeau M. (eds.) *Associated Regional Chronologies of the ancient Near East and the Eastern Mediterranean, Arcane Interregional*, Vol. III: Miscellaneous Materials. Leuven: Brepols.
4. Van de Velde T. Accepted for publication. Archaeometrical studies on finds: Bitumen. *In* Askar Chaverdi A. & Callieri P. (eds.) *From Palace to Town: Report on the multidisciplinary project carried out by the Iranian-Italian Joint Archaeological Mission at Persepolis, 2008-2012*. Oxford: Archaeopress.
5. Van de Velde T. Accepted for publication. Geochemical analysis on the bitumen lining of a vessel excavated from Tumulus B-5 by Peter B. Cornwall. *In* Porter B. & Boutin A. (eds.) *Embodying Ancient Dilmun: The Peter B. Cornwall Expedition to Bahrain and Saudi Arabia*. Boston: American School of Oriental Research Archaeological Report Series.

An overview of the structure of this thesis and the contents of each chapter is outlined here below:

Part 1 - Introducing the material & the research

Chapter 1 (*Introduction*) introduces the subject, establishes the research context and highlights the chosen approach for this research.

Chapter 2 (*Bitumen as a natural resource*) starts off with an introduction to bitumen and determines this quite specific material. Subsequently, the different sources of bitumen in the Near East are highlighted and explained how material from these seepages was worked into a mixture. A final subchapter handles the different types of usage of bitumen.

Chapter 3 (*Geochemical screening of archaeological bitumen*) introduces the geochemical methods used in this type of studies. Subchapter 3.2 is a status quaestionis of the research prior to my own and was published in the journal *Arabian Archaeology and Epigraphy* and co-authored by J. Connan. Following the state of research, the chapter sets further out by dismantling bitumen into its different fractions and explains the possibilities for specific chemical research. Finally, the methodology for the geochemical screening in this research is explained in detail.

Part 2 - Geochemical analysis on bitumen and its interpretations

All chapters in this part cover a specific bitumen dataset, covering multiple chronological periods, spread out over the entire Persian Gulf.

Chapter 4 (*Neolithic-period bitumen*) is the first chapter covering the analysis of a specific dataset. The dataset discussed in this chapter is a collection of bitumen from Dosariyah (Saudi-Arabia). The details of the study are published in *Journal of Archaeological Science* (chapter 4.1.) but are also subject to appear in the excavation report of the archaeological site. The results were used to further characterize Arabian Neolithic-period trade and the relation between Mesopotamia and the Gulf. This topic was presented at the *Seminar for Arabian Studies* and published in its proceedings (chapter 4.2).

Chapter 5 (*Bitumen in Dilmun*) presents the work on 3 different datasets from Bronze-Age period Dilmun sites. The first subchapter presents the work I conducted on a dataset from Tell F6 on Failaka. This work is accepted for publication in the excavation report entitled *Tell F6 on Failaka Island. Kuwaiti-Danish Excavations 208-2012* (eds. F. Højlund and A. Abu-Laban) and will be published by the Jutland Archaeological Society (see chapter 5.1). Two other datasets originate from several burial mounds on Bahrain, one of which contains samples excavated by the Danish archaeological mission (Moesgaard Museum) (chapter 5.2) whilst the other fits in with a publication project on materials excavated by Peter B. Cornwall and currently located in the Phoebe A. Hearst Museum of Anthropology (University of California, Berkeley, U.S.A.) (chapter 5.3). The former dataset is accepted for publication as book chapter in *The Rise of Kingship and the Early Dilmun State in Bahrain* (ed. S.T. Laursen, published by the Jutland Archaeological Society Publications), whilst the latter will appear as a chapter in *Embodying Ancient Dilmun: The Peter B. Cornwall Expedition to Bahrain and Saudi Arabia* (eds. Porter B. & Boutin A., to be published by ASOR).

Chapter 6 (*Hellenistic Period bitumen in the Gulf*) presents the data on several bitumen samples from various archaeological sites, more specifically from Mleiha, Dibba and ed-Dur, all of which are key-sites for the Gulf in this period. Although these results have been presented at the 2013 Seminar for Arabian Studies (London), they are published here for the first time.

Part 3 - From individual datasets to a broader archaeological perspective

Chapter 7 (*An updated overview of bitumen trade in the Gulf*) is a thorough update of what was written in chapter 3.2 and published in 2010 in *Arabian Archaeology and Epigraphy.* The discussion in this chapter follows the bitumen through time in the Persian Gulf and explains all of the changes in suppliers that were observed after geochemical analysis of archaeological samples, thus answering the main research questions. This overview incorporates all what is known concerning bitumen in the Persian Gulf and sets out to explain the specific role(s) of bitumen.

Chapter 8 (*Final conclusions and future perspectives*) This chapter highlights the major contributions to our knowledge on ancient bitumen through this research. Also several possibilities for future research are discussed.

Appendix I (*Bitumen from Achaemenid contexts*) is accepted to be published as a book chapter for the Tol-e Ajori archaeological project. It contain the study of bitumen from 2 sites located deep into the heartland of the Achaemenid dynasty: Sad-i Shahidabad and Tol-e Ajori. These 2 datasets, however, were the very first I was able to get my hands on and formed an excellent case-study to test my newly-acquired chemical skills. I feel that these analyses should also be presented in this work as they are the very first contributions on bitumen analyses from Achaemenid sites. Also do the datasets used in this part contains bitumen from a never-before observed seepage, the location of which remains unknown. The results from Sad-i Shahidabad have not yet been published, but are mentioned in the detailed book chapter I wrote on the dataset from Tol-e Ajori, which was accepted for publication for the excavation report (to be published by Archaeopress).

Appendi II (*Bitumen in the 3rd millennium of the Near East*) is a contribution Jacques Connan and I prepared for the Arcane Project (Interregional Studies Vol. III: Miscellaneous Materials) and illustrates bitumen in 3rd millennium Mesopotamia.

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List of abbreviations

22R	22R-17α(H),21β(H)-Homohopane (C_{31}) (chemical compound)
22S	22S-17α(H),21β(H)-Homohopane (C_{31}) (chemical compound)
31R	see 22R
C_{30}	C_{30}αβ-hopane (chemical compound)
DOS	Dosariyah (archaeological site)
EA-IRMS	Elemental analyser – Isotope-ratio mass spectrometry
GC-MS	Gas chromatography – mass spectrometry
GCRN	Gammacerane ($C_{30}H_{52}$) (chemical compound)
GCRN/31R	ratio between chemical compounds Gammacerane and 31R
GCRN/C_{30}	ratio between chemical compounds Gammacerane and C_{30}αβ-hopane
Oleanane	18α-oleanane ($C_{30}H_{52}$) (chemical compound)
OLN	see oleanane
OLN/C_{30}	ratio between chemical compounds oleanane and C_{30}αβ-hopane
Tm	17α(H)-22,29,30-trisnorhopane (C_{27}) (chemical compound)
Ts	18α(H)-22,29,30-trisnorneohopane (C_{27}) (chemical compound)
Ts/Tm	ratio between chemical compounds Ts and Tm

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Part 1- Introducing the material & the research

Chapter 1 Introduction

1.1 Introduction and aims of the research

It is practically impossible to mention the Near East without having to think about the oil or petroleum industry. It is estimated that 53 % of the world's oil reserves reside in the Near East, representing 66 % of the OPEC-countries in 2013[1] (Fantini, 2014). Needless to say, this natural resource has reshaped the area drastically on many levels: the black gold still dominates the international market and highly influences the worldwide economy. It attracted nations to become 'suddenly' interested in the Near East, made developments possible and nations rise, created jobs and opportunities, but it also led to the destruction of natural landscapes, pollution, and several (armed) conflicts resulting into cold-war-relationships between nations.

Contrary to common knowledge —that petroleum is only a product of the last two centuries—, the oil has always shaped the East; there is evidence of the (exemplary) usage of bitumen as early as the Palaeolithic Period (Boeda et al., 1996) and the material became increasingly popular in the 7th and 6th millennium B.C. on several Mesopotamian sites for local use mainly. But with the advent of the so-called Ubaid-expansion, bitumen became an important export material. Consequently, bitumen is found in the archaeological record of almost all Mesopotamian tells. Yet the material seldom was of great interest to early explorers of this region. The focus of the many large-scale excavations that were common in Iraq during the late 19th and early 20th century was on unearthing big, spectacular and valuable archaeologica such as reliefs, funerary objects, statues,... Bitumen, being a material with a strictly utilitarian value rather than an

[1] The Organization of the Petroleum Exporting Countries is a cooperation between twelve countries and acts as a regulator in the oil industry. The organization was established in 1960 in Baghdad (Iraq) and currently resides in Vienna (Austria). The twelve members are Saudi Arabia, Iran, Iraq, Kuwait, the U.A.E., Qatar, Venezuela, Libya, Nigeria, Algeria, Angola, and Ecuador.

ornamental one, easily escaped the eye of archaeologists. Nevertheless bitumen was present in many of these objects, many of which excavated in the 1920's by Sir Leonard Woolley from the "Royal Graves" at Ur. Many of the items unearthed from these graves consists partly of bitumen, mostly as an adhesive. For example, the shell figures on the Standard of Ur were glued with bitumen on the wooden box (see Figure 61). Beside this use it was also commonly attested in architecture as a mortar.

The first real comprehensive study on bitumen was by the hand of R.J. Forbes (1958) which focused not only on the different types of bitumen usage, but also on bitumen in the cuneiform texts, the composition of bitumen mixtures and the possible origins of the material. The geochemical techniques for sourcing bitumen were not yet developed at the time of Forbes' work, which left him only to propose suggestions as to where the Mesopotamians gathered their bitumen. Later scientific works confirmed many of his hypotheses (Connan, 1988, Connan and Deschesne, 2007). But due to the evolving archaeological excavation- and registration techniques on the one hand, and the development of a geochemical toolset for analysing bitumen on the other, it became possible to determine the geologic origin of archaeological bitumen. Changes in bitumen source were noted for single settlements throughout time (Connan and Nishiaki, 2003, Connan et al., 1996, Connan and Oates, in preparation), exchange networks have been identified (Schwartz and Hollander, 2008, Stein et al., 1999, Connan and Van de Velde, 2010) and bitumen from Mesopotamian seepages have been identified at many sites in the Persian Gulf.

1.2 Research aims and –questions

Despite the efforts of several researchers, our knowledge on bitumen exchange networks remains very fragmented. In 2010, Jacques Connan and I published an overview of our current knowledge on this very specific trade based mainly on data from geochemical analysis (Connan and Van de Velde, 2010) (see chapter 3.2). This was the most comprehensive overview of bitumen trade in the Near East at that time.

But generally, all of the work on archaeological bitumen was conducted from a site-point of view. Bitumen was excavated, sampled and sent for analysis; the results of which then framed into the archaeological site and the link between seepage and supplier defined. This is obviously a logical course to pursue and it gives extremely valuable information on that level. However, there's another point of view: that from the bitumen itself. By looking at the material itself; knowing where it's from and where it all ended up it gives us another perspective on the matter, one on a much greater scale than site-level.

Since I have been trained and educated in the archaeology of the Persian Gulf, this was to be my main point of focus. It also made sense to choose this particular region as almost all of the bitumen was imported from either Mesopotamia or southwest Iran; with some exceptions where more local raw materials were used. The Persian Gulf has always been an important corridor connecting people and cultures, more often than for the conquest of valuable materials and economic gain. A lot of materials changed hands at settlements in the Persian Gulf throughout its history, such as metal, wood, textiles, food and fodder, animals, but also bitumen (Weeks, 2003, Tengberg and Potts, 1999, Potts, 1990a, Ratnager, 1981, Potts, 1993b). Several of these materials are prone to disintegration and are (almost) completely deleted from the archaeological record by the sands of time, but bitumen is relatively stable and has good chances of surviving thousands of year in the dirt. Additionally, chemical analyses make it possible to pinpoint the place of origin of bitumen samples, to locate the seepage where this natural resource breached the earth's surface.

The primary question in my research concerns these origins. I wanted to know where the bitumen unearthed at archaeological sites came from, and why they ended up where they were uncovered thousands of years later. The initial aim of my research was to track bitumen throughout the archaeology of the Gulf, from the earliest periods (Arabian Neolithic) to the Hellenistic Period; and consequently identifying the sources of this material. Knowing the sources, would then make it possible to uncover the interregional human networks by which bituminous material was transported.

There are of course some related questions to the main questionnaire. The most evident ones concern the nature of the bitumen as found on archaeological sites. What was the bitumen used for? And if a differentiation in usage between sites and/or periods can be noted, to what can this be attributed?

As we will see later, bitumen was seldom used in its pure form but rather a mixture was made (see chapter 3.2). The first question that arises from this fact concerns the composition of these mixtures. What are the major components and can we identify the mineral fraction? Information on the latter can give clues on the place of fabrication of the mixtures. For example, if crushed seashell is found in combination with northern Iraqi bitumen, the mixture was not made at the place of extraction but rather on a site located close to a shoreline. The same goes for the presence of unique- or rare minerals. A detailed study of the bituminous mixtures requires the use of specific analysis (notably thin-sections and X-ray diffraction) and is beyond the scope of this research. However, macroscopic observations were made for every bitumen sample and peculiarities noted. In some cases, this was enough to (partly) answer some of the related research questions.

1.3 Approaching the bitumen

My methodology to approach the bitumen networks is twofold. First, I inventoried *WHERE* bitumen has been found. This was done through literature-research. All data was gathered in a database containing information on the level of the context and on the level of the site. If possible, the possible functions of the bitumen were also recorded.

The second part deals with the geochemical aspect of the research. Archaeological bitumen are commonly mixtures holding several fraction (see chapter 3.3), and my research focused not on the additives, but rather on the actual bitumen and on identifying their geological origin, thus the place where they were extracted.

I was able to obtain a bulk sample from Mleiha (Hellenistic-period site, Emirate of Sharjah, U.A.E) which I could use to experiment and test several methodologies before selecting the most efficient one. The selected methodology was then applied to archaeological samples from different contexts and sites. Although in the past bitumen was unearthed from many dig sites in the Gulf, much was not sampled or lost through the sands of time. Fortunately several researchers currently active in the Gulf have shown great interest in having their bitumen analysed and were keen on cooperating (see Table 1 and Figure 1).

Most of the bitumen found on Persian Gulf-sites is not indigenous to region. Evidently most of it was imported and part of the intercultural trading that has been taking place in the Persian Gulf starting from the Ubaid period. The results obtained through geochemical research made it possible to characterize this trade in bitumen and form hypotheses on its role throughout time. The specific methodology used in the research will be explained in chapter 3.4.

Table 1 Overview of all sampled sites and their dating.

Archaeological site	Dating	Samples #	Project
Tell F6	Early 2nd millennium	13	Kuwaiti-Danish excavations at Failaka
Dosariyah	5000-4500 B.C.	20	Dosariyah Archaeological Research Project
A'ali	2000-1700 B.C.	10	Collapse: the Eclipse of the East and the rise of the Dilmun State in Bahrain
Tumulus B-5	2000-1800 B.C.	1	The Dilmun Bioarchaeology Project
ed-Dur	1st-2nd century A.D.	3	Belgian Archaeological Mission at ed-Dur
Mleiha	1st-2nd century A.D.	1	Belgian Archaeological Mission at Mleiha
Dibba	1st-2nd century A.D.	1	Directorate of Antiquities, Sharjah, UAE
Tol-e Ajori	Early Achaemenid	5	Joint Iranian-Italian Mission at Persepolis
Sad-i Shahidabad	Early Achaemenid	2	Joint Iranian-French Archaeological Mission

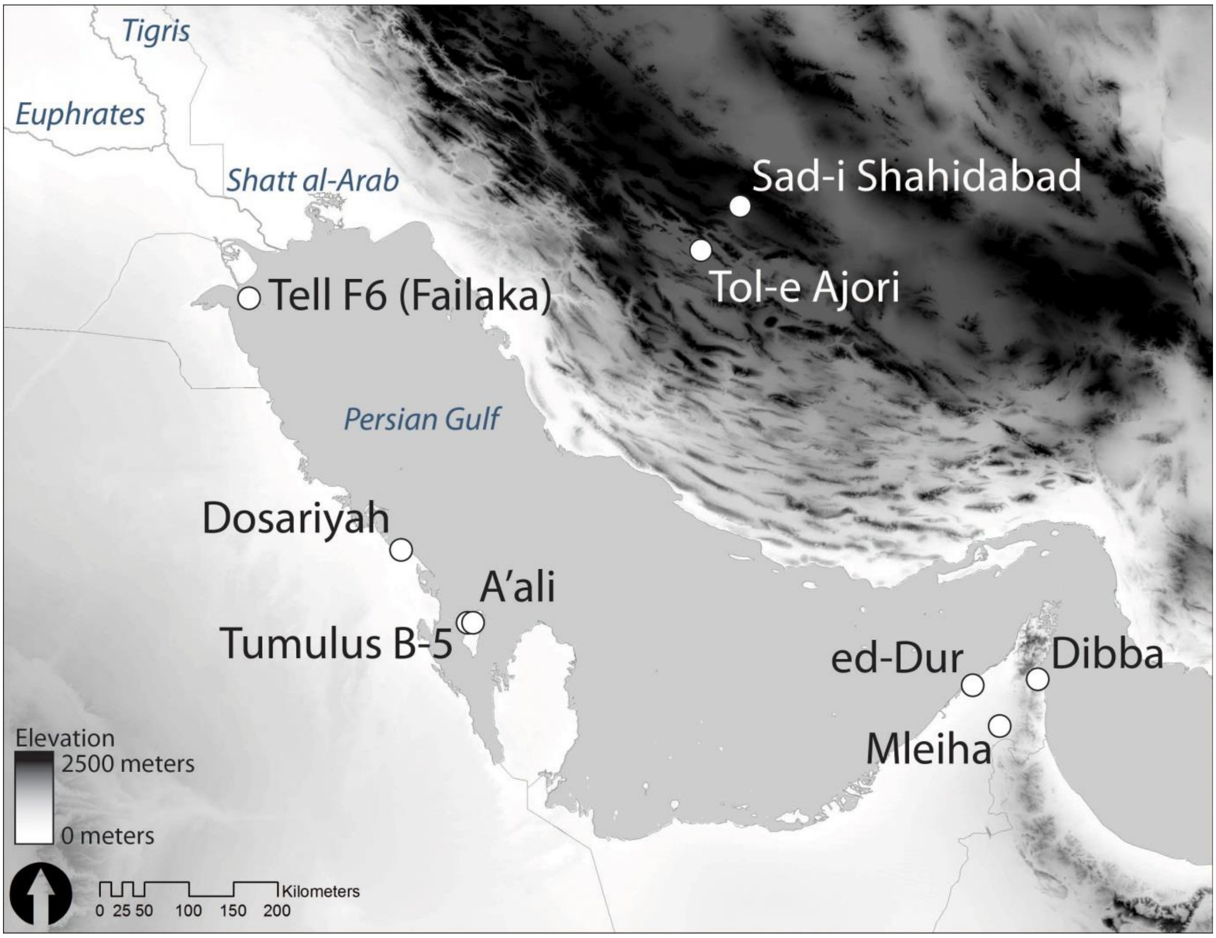

Figure 1 Map of all sampled sites used in this research.

Chapter 2 Bitumen as a Natural Resource

2.1 Introduction

It is practically impossible to mention the Near East without having to think about the oil or petroleum industry. It is estimated that 53 % of the world's oil reserves reside in the Near East, representing 66 % of the OPEC-countries in 2013[2] (Fantini, 2014). Needless to say, this natural resource has reshaped the area drastically on many levels: the black gold still dominates the international market and highly influences the worldwide economy. It attracted nations to become 'suddenly' interested in the Near East, made developments possible and nations rise, created jobs and opportunities, but it also led to the destruction of natural landscapes, pollution, and several (armed) conflicts and resulting into cold-war-relationships between nations.

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[2] The Organization of the Petroleum Exporting Countries is a cooperation between twelve countries and acts as a regulator in the oil industry. The organization was established in 1960 in Baghdad (Iraq) and currently resides in Vienna (Austria). The twelve members are Saudi Arabia, Iran, Iraq, Kuwait, the U.A.E., Qatar, Venezuela, Libya, Nigeria, Algeria, Angola, and Ecuador.

problem with organic material is obviously the decaying nature of the material and we have to rely often on either very little material evidence or rather on textual evidence. And although the bitumen also belongs in the category of natural resource/material, it stands the test of time much better than the above-mentioned products of trade. And like some other archaeological materials, it is also possible to determine the geological origin of bitumen. This gives vital clues on trading routes and spheres of interaction. With this information, we can start asking questions such as "*is there a change of bitumen origin through time?*", or "*why does settlement A gets bitumen seepage 1 whilst at the same time settlement B gets supplied from source 2?*", and most of all "*Why do we observe these changes in bitumen suppliers?*".

2.2 A Definition of Bitumen

Bitumen is commonly known for its usage in road construction and its application in roofing. It is mainly obtained through distillation of crude oil and represents the heaviest molecular fractions in crude oil (see Figure 2). During distillation, crude oil is evaporated at a temperature of 400° C and enters the distillation column. The evaporated fractions rise in the column and liquefy at different levels, depending on their molecular weight. The heavier fractions with a high carbon count will turn back to their liquid phase first, while the lighter fractions will rise higher in the column before liquefying.

Petroleum is the result of the exclusion of organic material (i.e. carbon) from the carbon cycle by deposition in a anaerobic contexts. Processes such as diagenesis and catagenesis may then transform the organic matter into petroleum (Peters et al., 2005a: 8). So in short, petroleum is the result of a transformation of dead organic materials. A decisive parameter in this transformation is the geological context. Three sort of 'rocks' need to be present for the formation- and preservation of petroleum:

- A source rock;
- A reservoir rock to hold the oil;
- A cap rock.

In the source rock, decayed organisms are transformed into hydrocarbons by the processes described above. Petroleum is still able to migrate through (rocky) sediments, unless there is a (impermeable) cap rock present, trapping the petroleum into reservoirs. This entire structure, including overburden rocks, is named a Petroleum System (see Figure 3). The geological formation of the cap rock is essential for oil or gas to accumulate into reservoirs. Whether it be fault traps, stratigraphic traps, or the formation of anticlines; a reservoir will form. The difference between petroleum and natural bitumen has to do with the geological circumstances during formation; unlike petroleum, natural bitumen does not migrate through geological rock formations and is indigenous to the source rock in which it was formed (Peters et al., 2005a: 360).

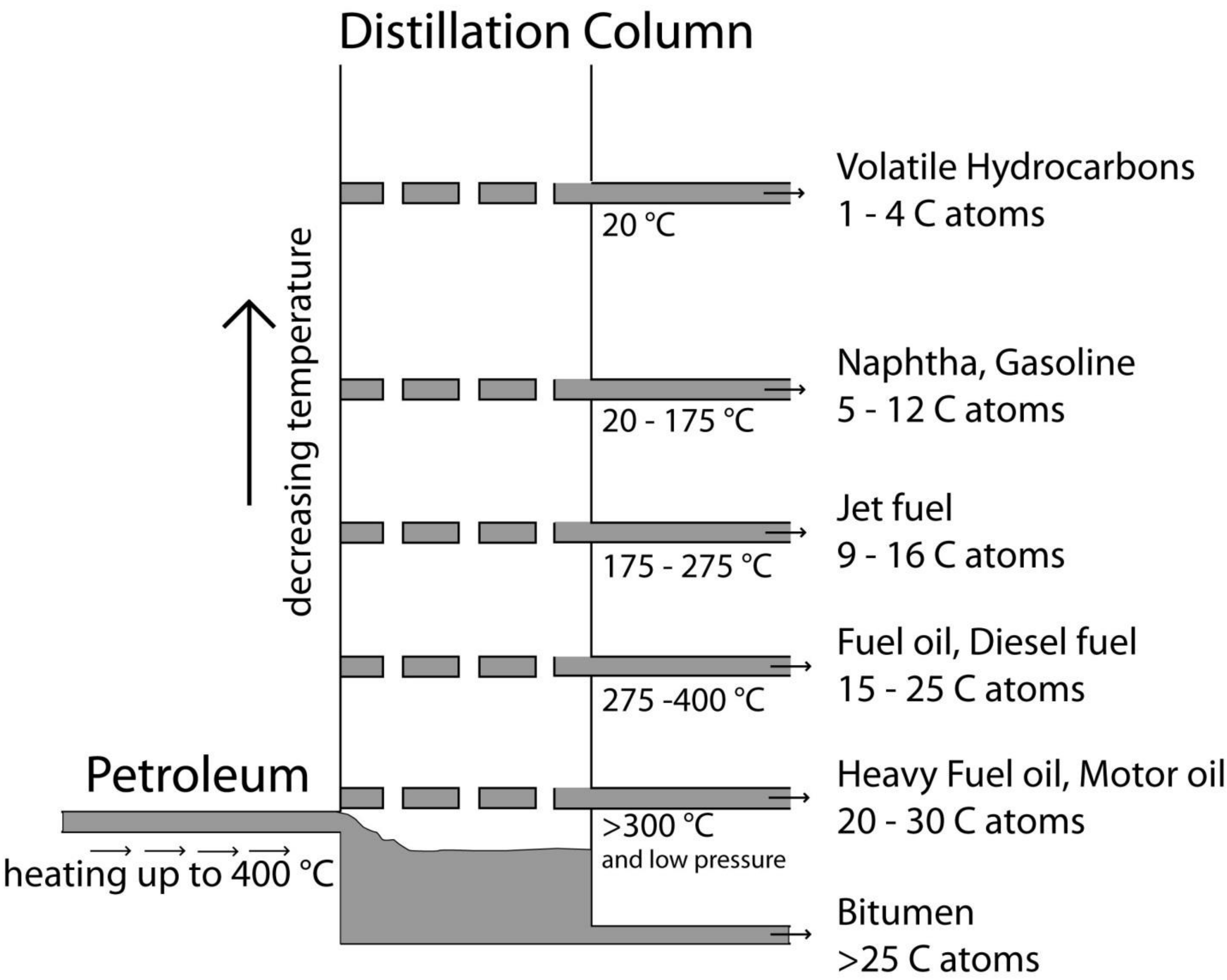

Figure 2 Simplified drawing of distillation column, as commonly used in the petroleum industry to separate petroleum fractions.

Many petroleum systems exist, but mostly they remain hidden with petroleum reservoirs, sealed off by a cap rock. When the cap rock is flawed, damaged, or subjected to geological faults, oil or bitumen from a reservoir can find its way to the surface. Surface seepages are much less common than the solid residues occurring in the rocks, nevertheless, they occur from northern Iraq to southeast Iran (Alsharhan and Nairn, 1997: 468). These seepages are visible at the surface, and have been used for both small- and large scale extractions of their resources.

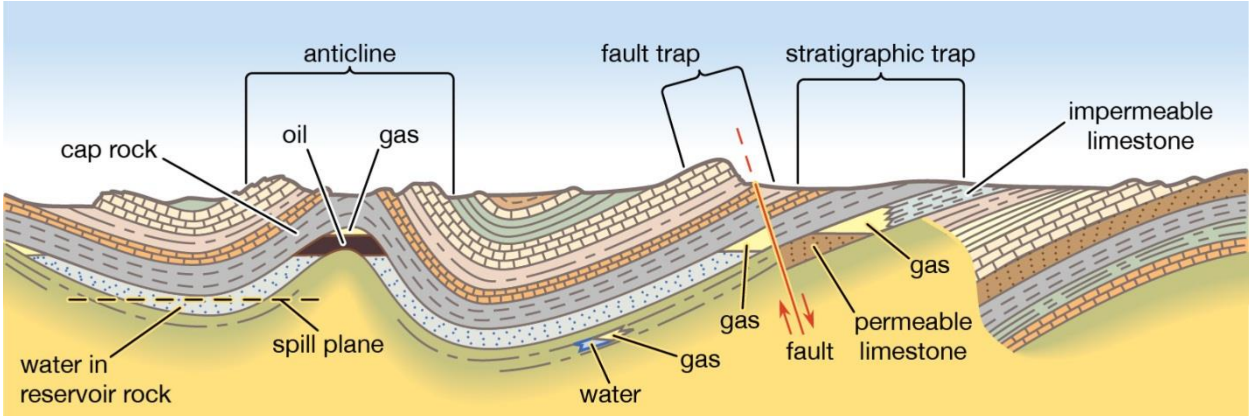

Figure 3 Schematized drawing of the different elements forming a petroleum system (image retrieved from the online Encyclopaedia Britannica, see (Riva, 2013) for bibliographic reference).Gas and oil are present in reservoir rocks, and may accumulate in stratigraphic traps due to presence of impermeable rock formations.

2.3 Bitumen in the Near East

2.3.1 Bitumen extraction sites

As mentioned above, bitumen seepages are a common occurrence in the Near East. An overview of all known seepages which are suspected to have been active in Antiquity are marked on Figure 4. Bitumen from most seepages have been identified at archaeological sites. From west to east, six major areas of bitumen extraction can be distinguished: the region of Ras Shamra, the Dead Sea, the Bichri-area in current-day Syria, Hit and the Mosul area in Iraq, and southwest Iran around the Deh Luran- and Susiana plains. For Iran, Mesopotamia and the Persian Gulf, only the last three areas are of importance and were major suppliers for ancient settlements (Forbes, 1964, Moorey, 1994, Connan, 2012, Connan and Van de Velde, 2010, Potts, 1997). Besides these key-areas, smaller, local seepages were also exploited. This was for instance the case for Demirköy Höyük which was supplied by the Bogazköy seepage (Connan et al., 2006a) and H3/as-Sabiyah in Kuwait where bitumen from the Burgan Hill was used (Connan, 2010, Connan et al., 2005). Remarkably, bitumen from neither Dukhan (Bahrain) nor Haushi (Oman) has been identified in archaeological samples. These seepages were probably not active during Antiquity or simply not known to the inhabitants of the area. The former option seems, considering the population and the size of the island of Bahrain, the most likely one for Jebel Dukhan.

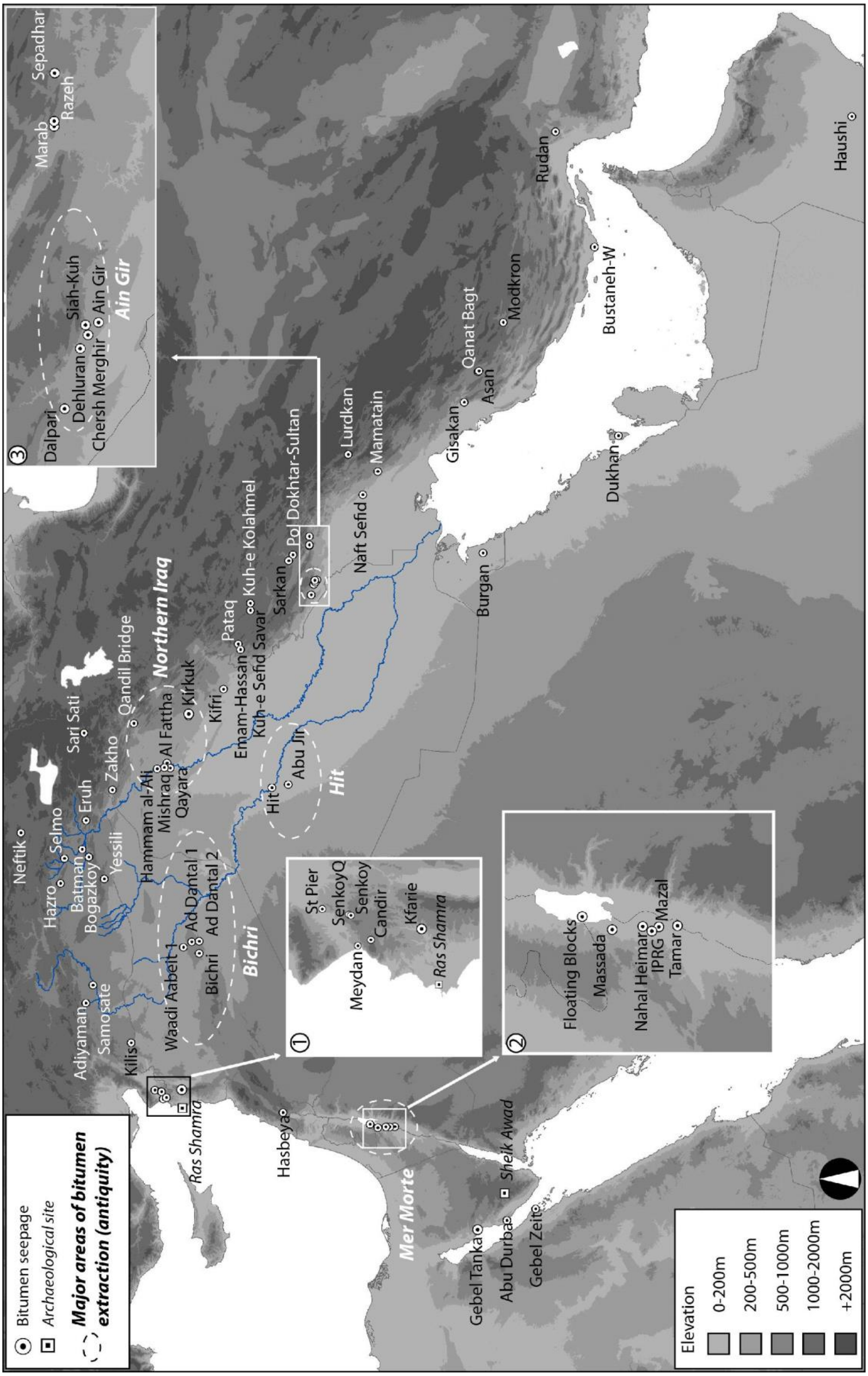

Figure 4 Overview of the known bitumen seepages in the Near East.

Especially important are the bitumen seepages of the Hit area (Iraq) that became famous in Antiquity and supplied a great many of settlements such as Terqa, Mari, Habuba Kabira, Kosak Shamali, Djebel Aruda, Tell Sheikh Hassan, Babylon, Tell el'Oueili in Mesopotamia proper (Connan, 2012: 27). Bitumen from this region has been identified not only in Mesopotamia but also multiple times in samples from sites in the Persian Gulf. The Hit-bitumen could reach the Gulf easily through networks and trading contacts due to the excellent location of the seepages alongside the river Euphrates providing plenty of opportunities for waterborne transport. These sources have been noted by Classical authors such as Vitruvius who wrote:

> "In Babylon there is a lake of considerable size that is called limnê asphaltatitis, "the asphalt marsh", and has liquid bitumen floating on its surface...". (Rowland and Noble Howe, 1999)

In Hit, bitumen surfaces through underwater seepages creating a film of bitumen on the water. After deposition on the shorelines it could be easily gathered (Connan and Deschesne, 1996: 18). A similar phenomenon is observed in the Dead Sea. Besides the Classical authors, there are cuneiform texts referring to these seepages, the most famous is one in which Hammurabi of Babylon expresses his 'wish' to Zimri-Lim of Mari:

> "The strength of your land are donkeys and wagons, and the strength of this land are ships. Exactly because of the [bitumen] I want this city" (Stol, 2012: 59).

As we will see later (see chapter 2.3.3.2), one of the major uses of bitumen is in naval architecture for which massive quantities of bitumen are required. Seemingly inexhaustible seepages supplying massive amounts of bitumen was obviously an interesting prospect for Hammurabi, especially considering that bitumen is one of the very few readily available natural resources in Mesopotamia.

Bitumen was easily gathered, and no specific installations were necessary, likewise for the creation of a bitumen mixture (see chapter 2.3.2). Consequently, this undoubtedly substantial industry often left little to no traces. In the literature, Tepe Farukhabad (Iran) has been identified as a unique example of a settlement controlling the bitumen trade, by extracting and processing bitumen prior to export (Wright and Berger, 1981, Badel, 2007). The site is located in the immediate vicinity of the Aïn Gir seepage, which supplies about 10 liters of pure bitumen daily and whose bitumen has effectively been identified archaeologically (Marschner et al., 1978: 98). There is, however, no clear evidence that Farukhabad had an exclusive right on the natural resource, nor that the bitumen mixtures were produced there. The data on which this idea was based, has not been backed up by results from geochemical analyses, consequently we cannot claim whether or not the site was so unique in its context (Connan, 2012: 137). Undoubtedly bitumen from the Aïn Gir seepage was used in Farukhabad (Marschner et al., 1978); however this bitumen has also been identified in other settlements, possibly with direct access to the seepage as well. Nevertheless, it remains very difficult to make any secure statements on the exact nature of bitumen extraction and on how this economy was organized.

2.3.2 Bitumen mixtures

Because bitumen has a tendency to turn into a (semi-) liquid state when exposed to heat, it was seldom used in its pure form. Therefore, almost all archaeological bitumen are mixtures of natural bitumen with a temper (Forbes, 1964: 56). Generally, mixtures consist of bitumen with mainly mineral matter (notably sand) and sometimes vegetal elements, with the actual bitumen accounting for 20- to 30 % of the total volume of the mixture (Connan and Deschesne, 1996: 117, Forbes, 1964: 56, Connan, 2012: 141).

Vegetal matter is not in all archaeological samples identified, and even differences in mixtures from the same dataset are attested such as illustrated in samples from H3/as-Sabiyah (Connan et al., 2005: 43, Connan, 2010: 269) and Kosak Shamali (Connan and Nishiaki, 2003). It is however hard to exactly assess this matter as the organic material originally present in the samples may have (extensively) weathered, as often illustrated by vegetal voids in bitumen pieces (Connan, 2010: 269). This makes it hard to correctly assess the role and quantity of the vegetal matter in ancient bitumen mixtures, although it is generally accepted that this fraction was deliberately added to most of the bitumen mixtures.

The mineral matter present in the bitumen consists mainly of carbonate minerals and quartz. Sometimes also feldspar, often associated with quartz, is identified. Carbonate minerals and quartz are both common and widespread minerals and seldom are any other minerals associated with bitumen mixtures. This indicates that no specific materials were used or necessary for the creation of bituminous mixtures. It is generally believed that the composition of the mineral matter is of no importance, and that ordinary sand was added to the heated bitumen until the desired viscosity was reached (Connan and Van de Velde, 2010).

The creation of the mixture is considered as a rather simple process for which no specific knowledge or craft was necessary. The bitumen was heated until it reached its melting point, which is around 160°C, after which the temper (vegetal and mineral matter) was added until the desired viscosity was reached (Connan and Deschesne, 1996: 89, Forbes, 1964: 63-66). Forbes (1964: 64) notes that the mixtures

> "...were undoubtedly prepared in earthenware jars or pots, not too big for handling. Curiously enough, I found in a Tell Asmar mastic [...] a potsherd of coarse earthenware [...], whereas the inside was rather porous and saturated with bitumen, a few particles of carbon still sticking to the inside. This not improbably a fragment of a pot used for the preparation of mastic and it is equally not

impossible, that some of the bitumen-lined jars, found in Mesopotamia, are really the actual melting pots of mastic.".

This observation is very valuable, and ever since the initial theory, many more pottery sherds with bitumen incrustations have been found such as at Tell Brak and Kosak Shamali in Mesopotamia (Connan and Oates, in preparation, Connan and Nishiaki, 2003), Tall-e Geser in Iran (Connan et al., 2014), and finally al-Khidr, Dibba and ed-Dur in the Persian Gulf (Belenová-Štolcová, 2010, Connan and Van de Velde, 2010, Van de Velde, personal observation). We should mention that these examples are not contemporary and are spread out over a large geographic area, which implies that the process of creating bitumen mixtures was a unified and unchanging practice.

Beside in earthenware vessels, bitumen mixtures could also be produced in ovens and even simple fire pits. The problem with the latter is that it is hard to identify this archaeologically. Previous research has shown that the repeated heating of bitumen causes them to become very brittle and lose their consistency due to the loss of entire compound classes (Hollander and Schwartz, 2000: 89). Such a firing pit is reported from RJ2/Ra's al-Jinz (Oman) (Cleuziou and Tosi, 2000: 36) and multiple installations were probably also present at Tell F6 (Failaka Island, Kuwait) (Calvet and Gachet, 1990, Van de Velde, Accepted for Publication-b). Considering the fact that bitumen mixtures were made inside an earthenware vessel and the decaying nature of bitumen —especially when fired for longer periods and at intervals—not much of these infrastructures are known. There is however one exception; the bitumen oven at Nippur (Iraq). This specific oven is a unique structure in the archaeological record and should be dated in the Early Dynastic II Period (McCown et al., 1978: 16). Undoubtedly, such installations were only used when large quantities of bitumen needed to be processed on-site.

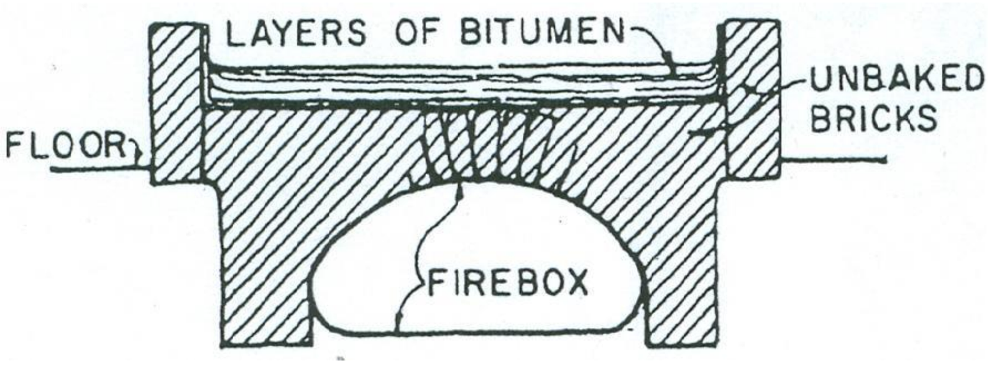

Figure 5 Section view of the EDII bitumen oven from Nippur (Iraq). Image source: McCown et al., 1978, Plate 42A.

A crucial issue in the interpretation of bitumen preparation is its frequent re-use. A major indication of reuse is often the location where bitumen is found on ancient settlements: not seldom they are found in one specific building or room functioning as a warehouse or storage. This phenomenon has been attested at Kosak Shamali in Mesopotamia (Connan et al., 2005: 35) and various sites in the Persian Gulf such as

H3/as-Sabiyah (Carter, 2010: 100-101), RJ2/Ra's al-Jinz (Cleuziou and Tosi, 2000: 30) and Umm an-Nar (Frifelt, 1995). Most bitumen at these locations bear marks of previous usage such as lashing, imprints of reed and ropes, or even barnacles.

2.3.3 Different types of usage

2.3.3.1 The common usage of bitumen

Bitumen was a key resource in the Ancient Near East and was used in many ways. The first usage of the material was as a glue to set flint fragments into handles, creating composite tools as sickles (Boeda et al., 2008, Boeda et al., 1996). In the same way bitumen can be found in many Mesopotamian pieces of art such as the Standard of Ur (see Figure 61), the Lyre of Ur, the famous Ur Ram Statuettes or the with mosaic-inlaid ostrich eggs (Aruz, 2003).

The waterproofing capacity of bitumen was also exploited extensively, especially in architecture. A bitumen coating could either be applied directly (such was the case for instance at Mari and the ritual bath at Mohendjo Daro) or by using reed mats. Mats coated with bitumen have been found on several sites, either as a flooring or shielding to waterproof roofs (Frifelt, 1995: 226). In some case, bitumen was applied directly on stone- or mudbrick courses as a mortar.

Bitumen was also used to seal pottery and other vessels. Many woven reed or palm leaves vessels, coated with bitumen on both sides in order to waterproof the vessel (Højlund, 1995: 101), are found in the Persian Gulf, Syria, Iraq and Iran. The density of these vessels at the Saar burial ground seems to indicate that they were embedded in the everyday life of Bronze Age at Bahrain Island (Moon, 2005: 196). Unfortunately, due to the perishable materials out of which these objects were made, many have been lost due to weathering or are in very poor condition, which makes them very hard to identify.

Many bitumen found on Persian Gulf-sites are shapeless lumps or small fragments with imprints, but one remarkable category of finds is often found as a real object, and that are the numerous stoppers/plugs. This type of artefact is introduced in the material culture of the Gulf at the same time as the introduction of actual bitumen. They have been identified at Dosariyah (n=15) (Drechsler, 2014) and probably one bitumen artefact from H3/as-Sabiyah can also be attributed to this object class (Carter, 2010: 97). These objects have been found frequently in Bronze Age levels at various archaeological sites such as Umm an-Nar (U.A.E.), al-Khidr (Failaka, Kuwait), Saar and Qala'at al-Bahrain (Bahrain) (Moon, 2005: 193, Frifelt, 1995: 226, Barta et al., 2008: 125, Højlund and Andersen, 1994: 408). These objects can appear in two different distinctive shapes. Type 1 is conical in shape and often topped with a thicker part or sort of knob. Type 2 is more elaborated and consist of a cylindrical end topped with a disc-shaped piece wherefore this type of objects can probably more be seen as a lid. These were manufactured to seal containers and were made to shape when the bitumen was still hot and mouldable.. To make a tight fit in their containers, probably also palm leaves were wrapped around the cylindrical part of the stopper (Moon, 2005: 193).

Figure 6 Bitumen stoppers from the two different types. The four on the left were excavated at Dosariyah (Saudi-Arabia) and represent the first type, whilst the one on the right was found at Saar (Bahrain) and belongs to type 2. Those from Dosariyah are courtesy of Philipp Drechsler, those from Saar have been published (Moon, 2005: 197).

2.3.3.2 Bitumen in naval architecture

There is evidence for the coating of boats with bitumen from the 6th millennium B.C. onwards, a practice that has been attested as late as the 20th century A.D. It not only makes the hull waterproof, but also protects it from mechanical damage (Connan et al., 2005: 22). Beside the 20th century parallels, the practice (or information relevant to it) is described in cuneiform texts, the most famous of which of course is the construction of a boat by Ut-napishtim in the Gilgamesh epic to escape the flood ordered to exterminate mankind. The practice of caulking boats with bitumen was considered very important and it was even regulated in the second millennium B.C. through the Codex of Hammurabi (§ 235):

> "If a boatman caulked a boat for a seignior and did not do his work well with the results that the boat sprung a leak in that very year, since it has developed a defect, the boatman shall dismantle that boat and strengthen it at his own expense and give the strengthened boat back to the owner of the boat." (Forbes, 1964: 91-92)

Quite clearly from this law, bitumen was a necessity in Bronze Age naval architecture,. Several cuneiform texts from the Ur III period have been studied in relation to both the construction of boats and the working of the dockyards that built the boats for the Dilmun- and Magan trade (Zarins, 2008, Potts, 1995, Carter, 2012). The exact amount of bitumen necessary for constructing a seagoing vessel has been subject to various calculations and is obviously dependant on the size of the ships. A text from Umma (TCL V: 5673) states that 6.12 tons of bitumen was necessary for the construction of one 120-gur ship[3] (Carter, 2012: 363, Zarins, 2008: 214).

The bitumen coating of ships was, however, not permanent. Ships often had to be re-coated with bitumen. Thesinger (1964: 124) accounts of such practices when he was among the people living in the south-Iraqi marshes in the 1950's:

> "[...] where his canoe was being recoated. The bitumen never lasted more than a year, after which it began to crack and let in water. These cracks could be temporarily sealed by heating the bitumen with a torch of reeds."

Anthropological studies revealed that the vessels in the 20th century marshes of Iraq were yearly stripped of their bitumen by hammer and chisel, after which the wooden frame of the boat was repaired and recoated. This recoating (see Figure 7) is done by heating the bitumen, whether or not with the addition of new bituminous material, and reapplying it to the hull (Ochsenschlager, 1992: 52). We can assume that the 3rd millennium B.C. boats also needed the same sort of maintenance, perhaps even more considering the rougher conditions at sea as compared to the still waters in marshlands.

As of yet, no archaeological evidence of wharfs has been found and we have to rely solely on the information from cuneiform tablets and anthropological data for the Mesopotamian part. However, several bitumen found at sites in the Gulf are clearly stripped from the hulls of boats —the evidence of which lies in the presence of barnacles on the bitumen or imprints of rope/planking—, the most elaborate cases being excavated at Ra's al-Jinz and H3/as-Sabiyah (Carter, 2010, Connan et al., 2005, Carter, 2006, Vosmer, 2000, Cleuziou and Tosi, 1994).

[3] The **gur** is a unit of measure and is commonly accepted to describe the capacity rather than the boats itself. One gur is thought to be the equivalent of 300 liters.

Figure 7 The recoating of a *tarada*, a traditional planked watercraft of the Iraqi marshes, these boats measure about 11 meters long (Thesiger, 1964: plate 74).

Chapter 3 The Geochemical screening of Archaeological bitumen

3.1 Introduction to this chapter

Several aspects of the bitumen research will be tackled in this chapter. The first subchapter (3.2) is a status quaestionis of the knowledge on bitumen trade based upon the results from geochemical studies prior to my own research. In a way, Chapter 7 is an updated version of this text based upon the analyses that were conducted for this research. This chapter further sets out to explain some basics on bitumen and the fractions it contains, providing all necessary information to continue to Chapter 3.4, where the handled methodology for chemical analysis on archaeological bitumen is explained in detail.

3.2 Status Quaestionis

The content of this chapter has been published in :
Arabian archaeology and epigraphy

Connan J. & Van de Velde T. 2010. An overview of bitumen trade in the Near East from the Neolithic (c.8000 B.C.) to the Early Islamic Period. *Arabian archaeology and epigraphy, 21:1, 1-19*

3.2.1 Introduction

Since the pioneering studies of Forbes (Forbes, 1964), Marschner *et al.* (Marschner et al., 1978) and follow-up investigations by Connan (Connan, 1988), the results of numerous analyses have been gathered, step-by-step, at various archaeological sites in Iran, Iraq, Israel, Turkey, Oman and the Gulf. These results have been partly released in excavation reports and papers, or published in peer review journals. A significant volume of dispersed information, covering both a wide geographic area and also a large time scale, is therefore presently available, but it is sometime difficult to access as it is for not always found by electronic tools.

This paper has been undertaken as a follow-up to the Master Thesis of one of us (Thomas Van de Velde), which was devoted to the origin of bitumen in excavations from the Near East. In this university work, elaborated in close cooperation with the second author, published data were compiled, leading to conclusions about the bitumen trade in the Near East through time. This basic synthesis served as the starting primary point of this paper, which has been updated and enriched with new information sorted out from files of ongoing and unpublished studies carried out by Jacques Connan. Therefore this article forms an excellent status questionis of the research topic prior to the analysis of new datasets.

The aim of the chapter is to outline some prominent features of the trade of bitumen in the Near East, from the very early days of the Palaeolithic to the Early Islamic period. We are conscious that this overview only reflects the present state of research; any additional study will upgrade the story and refines the pattern of exchange links, and might modify the ideas on the subject. Nevertheless the amount of data, which cover more than 2000 samples of bituminous mixtures analysed over more than 20 years, allows us to propose already some general statements which we consider valuable enough to be reported.

3.2.2 The importance of local sources

It is quite logical to assume that settlements did not import bitumen unless necessary. Importing goods always brings along an extra cost: the products have to be transported, which requires appropriate means of transportation and middlemen, who must be paid. One can assume that this statement was valid in Antiquity as well, and that there was a preference, if possible, for using local raw materials. Analyses of bitumen at many archaeological sites in the Near East confirm this assumption and indicate that it was a common practice at any time and any place: from the Palaeolithic period at Umm el Tlel in Syria (ca. 70.000 BP) to the Bronze Age at F6 on the Failaka Island. This practice however, even if logical, is not observed everywhere. For instance the oil seeps of Bahrain at Djebel Dukhan have not been identified in the archaeological bituminous mixtures analysed so far on the island (Connan et al., 1998). This feature may be due to several reasons:

- either the most relevant samples, i.e., the oldest ones which may include local bitumen, are not yet analysed.
- or the local bitumen, collected in limited amounts, was diluted with imported bitumen and is no longer detectable;
- or bitumen users relied on a more abundant supply, easily and regularly imported from abroad, i.e., from Mesopotamia or Elam. Local seeps, which had obviously low yield and where it was difficult to gather the bitumen, may not have been used for it was easier to get bitumen from more distant sources;
- or different qualities of bitumen were required and the Jebel Dukhan bitumen may have not been suitable to be used for whatever purposes bitumen was needed.

Site F6 on Failaka Island did not only bear Hit bitumen, but one out of the ten samples analysed had an unmistakable Burgan Hill provenance, the bitumen source closest to the island. The samples taken are all situated in the Early Dilmun period (ca. 2200 – 1700 BC), a period in which Failaka was a major player in the Persian Gulf trade; evidence of both Mesopotamian and Dilmunite presence (Potts, 1990a: 291) suggests that the island fulfilled an important role in the trade between those two regions. The special bond that Failaka Island shared with both Dilmun and Mesopotamia resulted in a steady flow of imported bitumen, as we will see later, almost entirely with a Hit provenance (similar to what happening at Bahrain at that moment, cf. infra). Bahrain Island, however, had a more complex situation with Iranian sources used outside Qala'at al-Bahrain. The occurrence of Burgan Hill bitumen during that time shows that this source was still in use, most likely supplying the sites on the shores of present-day Kuwait, but most importantly that this bitumen could also have supplied Failaka whenever needed. The bulk of bitumen was probably imported from Mesopotamia, as our samples point out, but in times of shortage the Burgan Hill bitumen could have been exported to Failaka Island, since this was the closest source. This bitumen, which in fact occurs as oil-stained sandstones, may have also been mixed with some imported bitumen for instance from Hit, to generate a bituminous mixture with the required properties, i.e. between 20 and 50 % bitumen. The input of Burgan Hill tar sand should bring much quartz into the mixture. The chemical signature of the composite bituminous mixtures, accessible today by analysis, may obscure the Burgan Hill occurrence if its contribution was limited. Such a situation is very likely, for quartz was not identified as the dominant inorganic constituent among the minerals present. In addition, the fact that the Burgan Hill tar sands contain only around 10% bitumen probably did not make them the preferred bitumen raw material during the mid-third millennium and later: most contemporary mixtures analysed contains between 15 and 50% bitumen as recorded at many archaeological sites. However imported mixtures with lower concentration of bitumen (18 and 12%) were also prepared, as seen recently at Dibba (U.A.E.) in 1st century A.D. storages jars filled with bituminous mixtures (Figure 8).

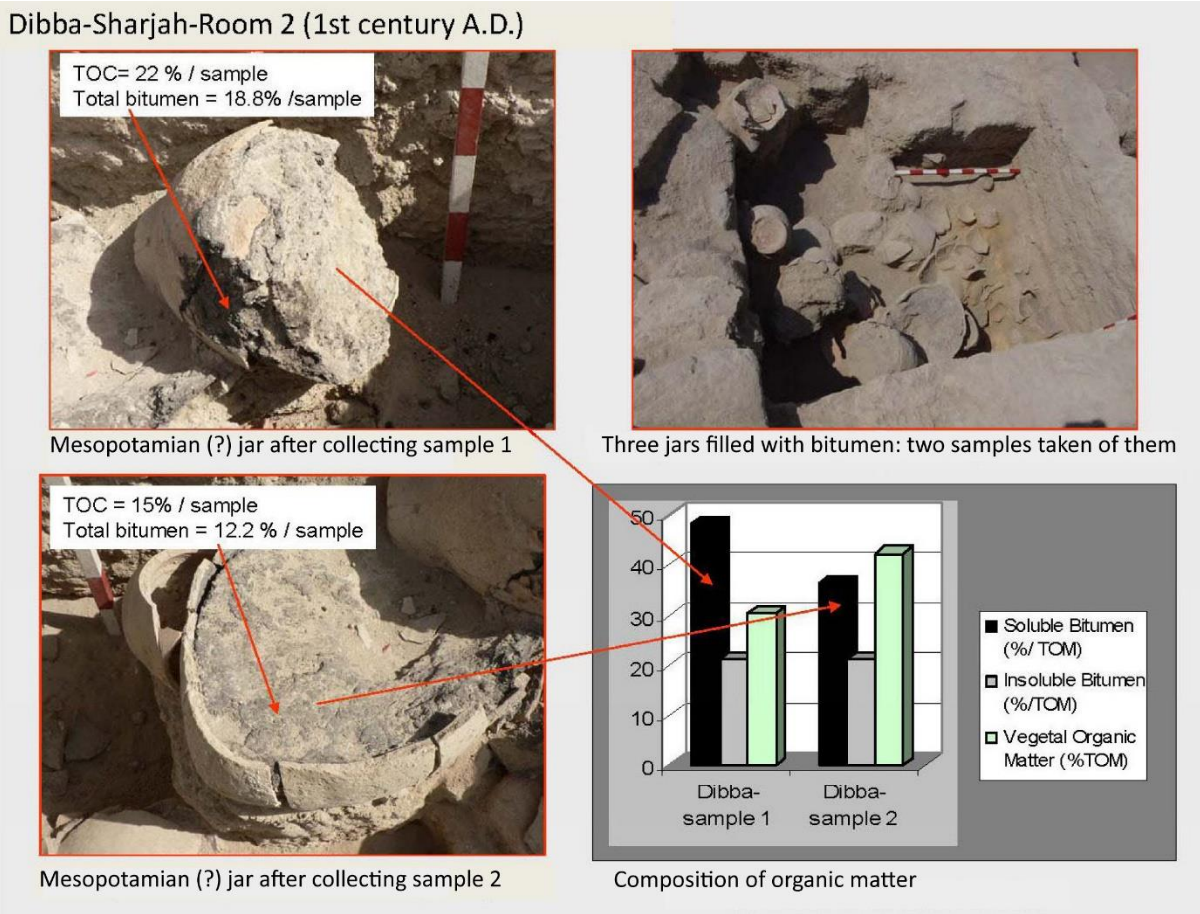

Figure 8 Picture (©dr. Sabah Jasim) of the bituminous mixtures discovered in the first century A.D. jars of room 2 during excavations at Dibba (Sharjah, U.A.E.).

Consequently small springs of oil were probably actively exploited, especially in areas where the bitumen quality is good, but they may have been abandoned when a steady flow of foreign raw materials was insured through stable trade routes. This imported bitumen could then be easily bought at open markets and stored.

In Iran, the inhabitants of Tall-e Abu Chizan site got their bitumen from a local source, likely Naft Safid oil seepages, during the fifth millennium BC (Connan et al., 2008) while during the same period, bitumen from Masjid-i-Suleiman and related sources were exploited for sites located in the Susiana plain (Marschner et al., 1978: 110). Naft Safid oil seeps are the closest sources of bitumen for Abu Chizan settlements and were therefore used preferentially by their inhabitants.

Gregg *et al.* documented another case history in Iran with bitumen analyses in pottery vessels from earliest agricultural villages and pastoral encampments at Ali Kosh, Chagha Sefid (6800-7200 BC) and Tepe Tula'i (6200-5900 BC) (Gregg et al., 2007). If a perfect match between archaeological samples and the analysed Chersh Mehrghir oil seep was not reached, it is very likely that the source of many bitumen belong to the area of Deh Luran, Ain Gir, Siah Kuh, Chersh Mehrghir. However bitumen from Tepe Tula'i are more diversified, which hints to the fact that they were imported from several sources including the Chersh Mehrgir area. Analysis of bitumen from another proxis, i.e., Tepe Aliabad site did not provide either a perfect match with the most likely source which is again Ain Gir-Siah Kuh. Such discrepancies may be due to differences in degree of alteration of steranes and terpanes between the present day references and their archaeological counterparts or simply means that the Tepe Aliabad bituminous artefact itself was imported as a manufactured object from Mesopotamia.

In Turkey archaeologists have found bitumen artefacts dating 8100 BC at the Neolithic site of Demirköy Höyük (Connan et al., 2006a). According to analyses carried out on artefacts, it was established that the bitumen used was likely collected in the close vicinity of the site. Though not providing a 100% match, the archaeological samples are very similar to the Boğazköy oil seeps (Figure 9). Perhaps the bitumen originates from another close-by source, or has been altered due to weathering. It is possible as well that the chemical properties of the oils exploited at Neolithic time were slightly different and that this limited source has been definitely exhausted.

Umm el Tlel, located in Syria, is a Middle Palaeolithic site where many worked flints were excavated, bearing some traces of bitumen in both the 40.000 BP and 71.000 BP layers (Boeda et al., 2008, Boeda et al., 1996).The inhabitants of the site exploited oil-stained quartz sands from the Bichri Mountains, located at about 40 km east of the site (Figure 9).

The coastal Neolithic site of H3, As-Sabiyah (Kuwait), dated to the late sixth to early fifth millennium BC (Ubaid 3), provided fifteen lumps of bituminous mixtures from reed boats, which after analyses, were given a provenance (Connan et al., 2005). The Burgan Hill oil-stained sands, located onshore in present day Kuwait, at 70 km south of As-Sabiyah as the crow flies, supplied the Ubaid-period site and were probably also used to glue the flint tools collected at Burgan Hill itself (Figure 10). This last assumption has not been cross-checked for the presumed bitumen remains on flint implements may have likely been altered and contaminated by soot during the burning of the Kuwait National Museum during the first Gulf War.

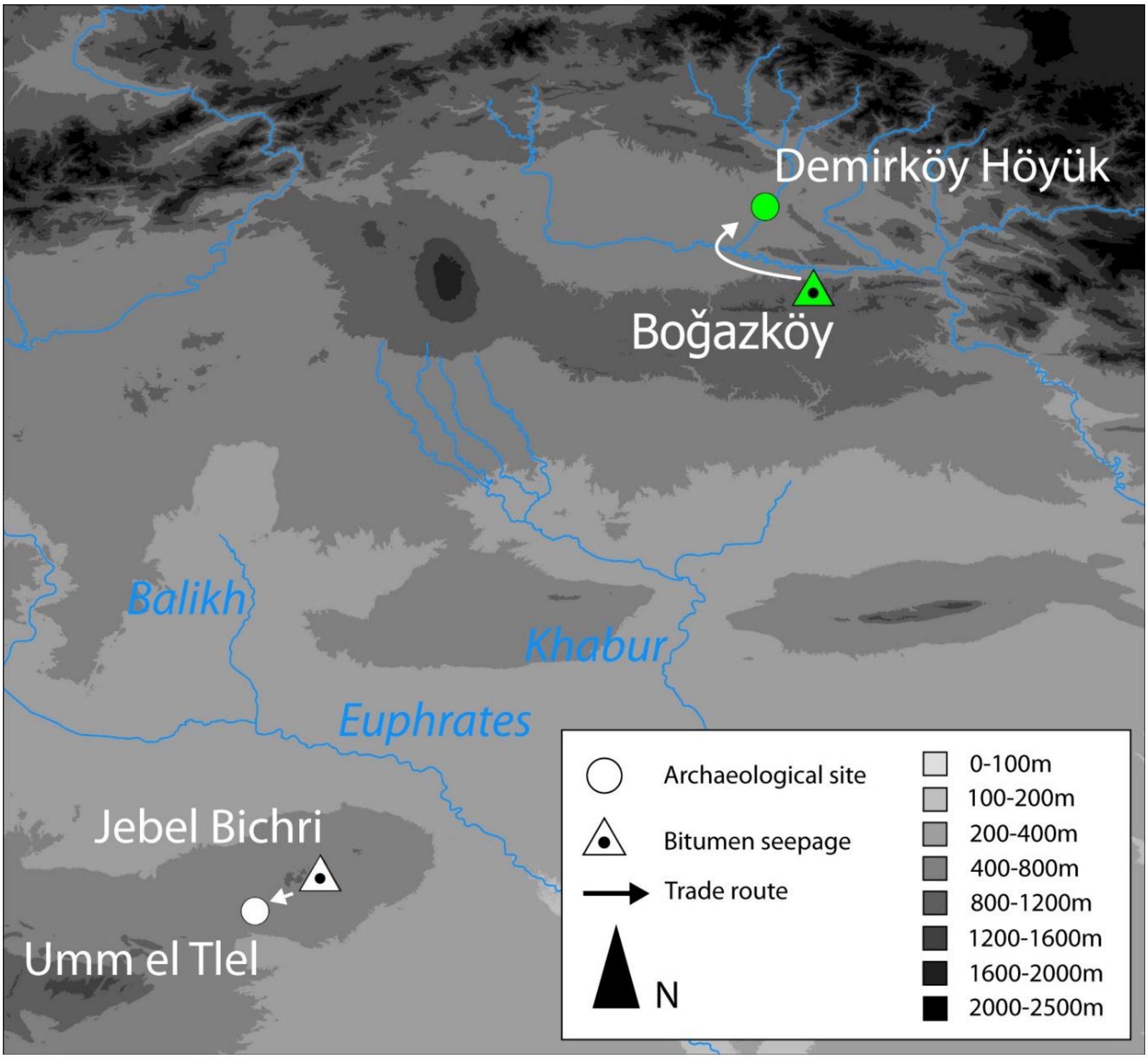

Figure 9 Early bitumen exploitation at the Middle Palaeolithic site of Umm el Tlel (Syria) and the Neolithic site of Demirköy Höyük (Turkey).

3.2.3 Early development in bitumen trade

3.2.3.1 From Ubaid 0 (late seventh millennium) to Ubaid 2 (mid sixth millennium)

From the Ubaid 0 (late seventh millennium) to the Ubaid 2 (mid sixth millennium) period, bitumen analyses are published from three sites only: Tell el'Oueili (Iraq), Chogha Mish (Iran) and Tell Sabi Abyad (Syria). Data collected from these sites points out that people were importing foreign materials. The inhabitants of Tell Sabi Abyad (6100-6200 BC) imported their bitumen from northern Iraq (Figure 10) and from an unknown source likely in southern Turkey (Connan et al., 2004) whereas at Tell el'Oueili we have a more complex story due to the availability of a complete bitumen sequence covering many periods. From the Ubaid 0 to Ubaid 2 periods, the settlement imported bitumen from northwest Iran. These Iranian sources supplied Chogha Mish (6800 BC to ca. 3200 BC., samples taken from layers situated ca. 6000 BC and 3200 BC) as well. This feature is in perfect agreement with general views on the Early Ubaid periods in Mesopotamia: most of the economic transactions and contacts were situated alongside an east – west orientated axis (Akkermans and Schwartz, 2003: 154, Huot, 1994)

3.2.3.2 Ubaid 3 to 5 (ca. 5300 – 3800 BC)

A major change in bitumen trade in Mesopotamia is noticeable starting at the Ubaid 3 period around 5300 BC[4] (Connan and Ourisson, 1993): the above mentioned east-west axis is now being re-orientated and runs north-south, alongside the Tigris & Euphrates rivers, and the Persian Gulf. As a result, Mesopotamian Ubaid products and materials are found as far as Anatolia in the north and the Persian Gulf in the West (Akkermans and Schwartz, 2003: 154). As a consequence, Tell el'Oueili was supplied with bitumen from northern Iraq (Connan et al., 1996, Connan and Ourisson, 1993) within the Ubaid 3 and 4 periods (between ca. 5300 and 4300 BC). Tell el'Oueili is not the only Mesopotamian site from the Ubaid period which provided bitumen samples; analyses have pointed out that Tell es-Sawwan (ca. 5500 – 5000 BC) imported bitumen from northern Iraq as well (Connan and Deschesne, 1992b, Connan and Deschesne, 1992a). The spread of this bitumen is not restricted to Mesopotamia; Ain as-Sayh (located in Saudi Arabia, ca. 4500 – 4000 BC) and Ra's al-Hamra (Oman, ca. 4400 – 3500 BC) imported their bitumen from the Mosul area as well (Connan et al., 2005, Connan and Carter, 2007) (see Figure 10). This is in perfect alignment with the above stated remark about the reorganization of long-distance trade.

It is rather doubtful though that there was an actual maritime trade network of bitumen from Mesopotamia to sites in the Persian Gulf. Mesopotamian painted Ubaid pottery, which along with bitumen is a clear mark of foreign contact, is common at Neolithic sites in the Eastern province of Saudi Arabia and at H3 in Kuwait. There were definitely contacts between Mesopotamians and the inhabitants of settlements on the shores of the Persian Gulf, and long-lasting maritime trading network existed in the Persian Gulf based on Ubaid pottery (Carter, 2006) but bitumen seems not to have been a major item of trade. Possibly, bitumen was also traded by these early traders, since it was a necessity in seafaring: bitumen was used for caulking the hull of reed boats, which makes them stronger against any damage and waterproof. An excellent example of this practice has been found at the site of As-Sabiyah, where archaeologists have unearthed

[4] This is the date as proposed by Forest (1996) for the start of the Mesopotamian Ubaid 3 period. We are aware of the difficulties of chronology for this period and the different suggestions for an absolute dating. Covering an area as large as Mesopotamia to the Persian Gulf makes the chronology even more complex.

bitumen slabs with clear imprints from boats (Carter, 2010). It is possible that the bitumen fragments found at the site of Ain as-Sayh can also be linked with the re-caulking of boats and the site thus could have been used as a strategically-placed harbour for Mesopotamian seafarers (McClure and Al-Shaikh, 1993). We must not forget that settlements on the shores of the Persian Gulf were not only places where goods could be traded, but they could also function as intermediate station to re-supply and, if necessary, repair boats (Ratnager, 2004: 230-231). Unfortunately, at the present state of research it is too uncertain to raise such assumptions, but it is definitely possible that similar sites existed.

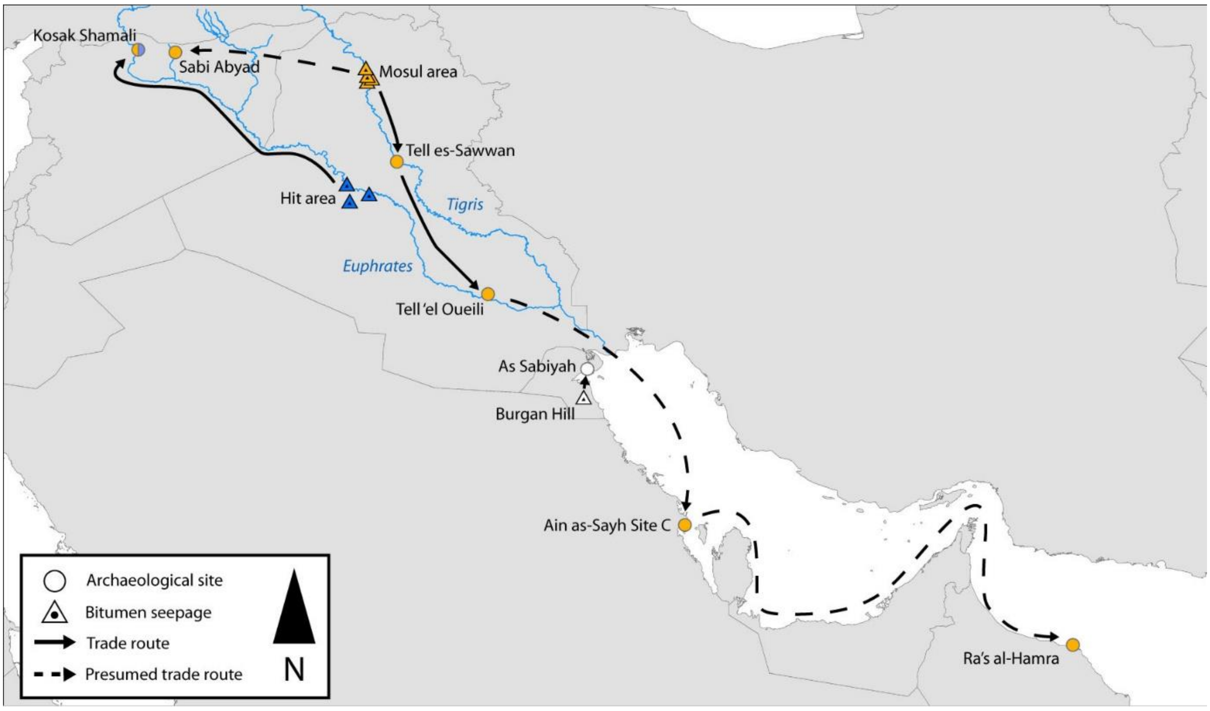

Figure 10 The exportation and trade routes of Iraqi bitumen in Mesopotamia and the Persian Gulf during the Ubaid Periods.

Though an unmistakable and remarkable trend, we must keep in mind other bitumen sources. Local sources still played an important part. Starting from this period we have evidence of the exploitation of the Hit bitumen, which was to become very important in the long-distance bitumen trade in later periods. At the Kosak Shamali warehouse, archaeologists have discovered bitumen which had 2 origins: Mosul and the Hit area (Connan and Nishiaki, 2003).

3.2.4 Uruk in Mesopotamia

The bitumen pattern at Tell el'Oueili changed again in the Uruk VI period (3500-3200 BC) and now bitumen from Hit only reached the settlement. From the Uruk period onwards, Hit bitumen became very common in Mesopotamia, especially at sites located along the Euphrates. Bitumen from these seepages has also been identified at Hacinebi Tepe (Stein et al., 1999) and Djebel Aruda (3200 BC, Connan unpublished) (see Figure 11). The question we must raise here is: "Why did the inhabitants of Tell el'Oueili, and probably those of other sites in Sumer, choose another bitumen supplier?" The answer to this question might lay in changing social structures and its direct consequences. The fourth millennium is a true turning point for Mesopotamian history: the introduction of the city and urban life. Why the changes from rural to urban life took place at this exact time and place is still a much debated subject, but the changes it brought to Mesopotamia are of uttermost importance. The birth of cities is also the birth of the first monumental architecture, which needed massive amounts of building materials, notably mudbrick and bitumen. Moorey (Moorey, 1994: 335) concluded in his work on Mesopotamian materials that, from the mid-fourth millennium, bitumen was used much more in architecture than before. This increased consumption can thus be explained by the rise of monumental architecture and the indissolubly growing demand for raw materials. This greater demand for raw materials must have led to larger scale exploitation which was possible at Hit, a region of exploitation that became more important at that time. In addition it seems that all along the Euphrates, upstream the city of Uruk, there was set a series of trade settlements with Uruk merchants who imported their technologies and practices and among them the use of bitumen. This was apparently the case at Hacinebi Tepe (Stein et al., 1999) where utilization of bitumen was generalized from the Uruk period, but also in other cities like Djebel Aruda (3200 BC), Habuba Kabira (around 3500 BC), Tell Brak, and Tell Sheikh Hassan (around 3500 BC). Though bitumen from Hit was already used earlier (from the Early Ubaid to the post Ubaid, 4800-4200 BC,), it seems that its use became generalized only from the Uruk period onwards. Why else would the inhabitants of Tell el'Oueili, who lived closer to the sources of Hit than those of northern Iraq, wait until the Uruk period to import their bitumen from this area? One explanation may be the establishment of a trade network all along the Euphrates from this period onwards. Material that is distinctively 'Uruk' in nature is to be found at many sites in the northern plains of Mesopotamia (Algaze, 1989: 577). Algaze defines these settlements as enclaves, stations, or outposts of the Uruk culture promoted by Uruk, the leading city in Mesopotamia. Whether or not this was the case, is rather hard to prove at the present state of research; Uruk might not have been the only major city in Mesopotamia where developments towards urban life were present, nor is it certain that the city of Uruk 'controlled' the rest of Mesopotamia. Despite these remarks; there is undeniably Uruk material present at sites in the northern alluvium, so contacts and a trade network must have existed, but we simply cannot say convincingly if this network was controlled by Uruk, or any other city in the alluvial plain, or if it was the result of merchants trading abroad.

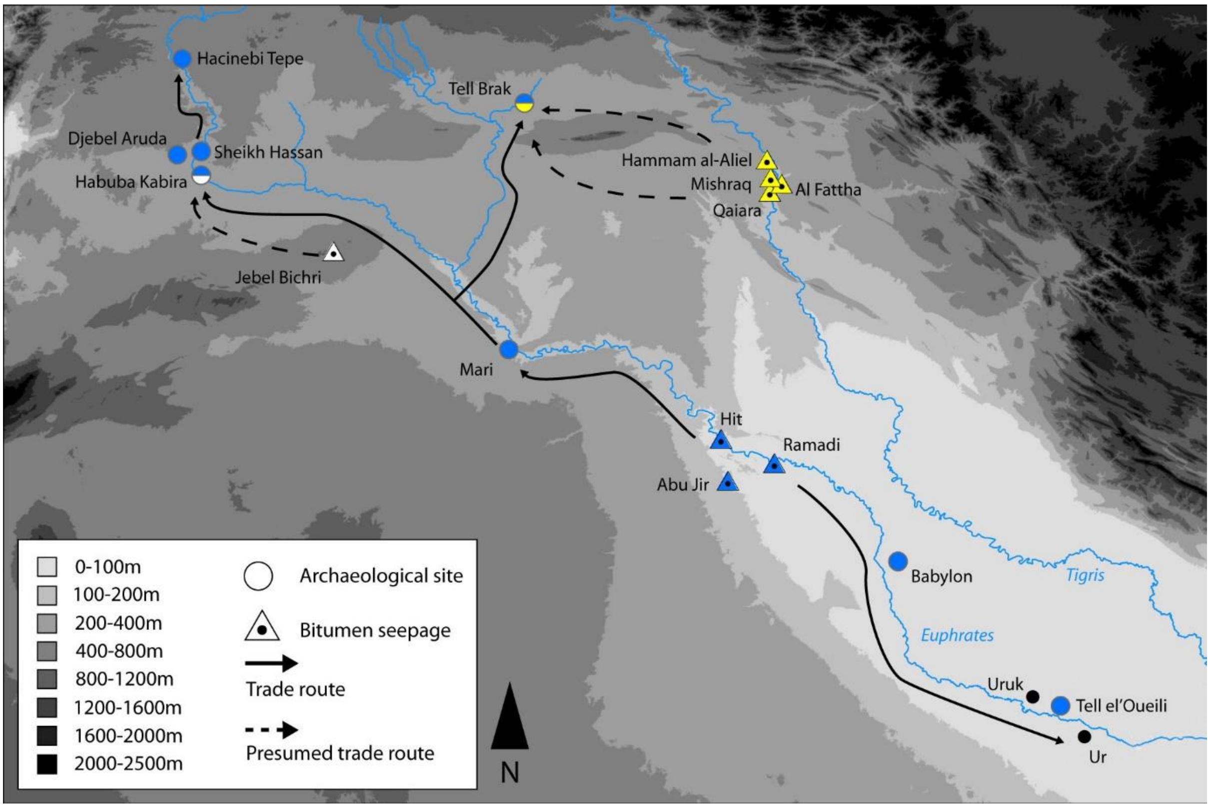

Figure 11 Bitumen trade routes in the Uruk- and later periods. Noticeable is the presence of a trade network along the river Euphrates for Hit bitumen. We also see the appearances of Jebel Bichri bitumen at Habubab Kabira, indicating the importance of local sources. Analyses of bitumen samples from Tell Brak points out that the Mosul area was still supplying settlements with bitumen. Quite possibly, this type of bitumen was more used at sites in the north of Mesopotamia alongside the river Tigris.

Tell Brak, located in northern Syria at the border with Turkey, imported its bitumen from both Hit and Mosul (Connan and Oates, in preparation). Bitumen from both areas reached Tell Brak in the Uruk period, but during the Bronze Age only northern Iraqi bitumen is found at the site. Unfortunately, only thirteen samples have been investigated so far, so it is dangerous to reach a decisive conclusion on this matter. But it seems, however, that cities of Assur region were supplied mainly by the Mosul area while those of Sumer got their bitumen from the Hit area.

3.2.5 Some trade routes at the turn of the second millennium B.C.

Texts from the Ur III period (2047-1940 BC) state that the province of Kimash and Madga, situated in the area of Kirkuk and Nuzi, delivered huge amounts of bitumen to the royal city of Ur (Connan and Deschesne, 1996, Connan and Deschesne, 1992a). At a first glance it might be surprising to note that bitumen travelled all the way down to the city of Ur in Sumer, whereas both Ur and the bitumen source of Hit and Ur, are situated alongside the Euphrates which provided easy transport. Nevertheless one must remember that many goods were brought down to Sumerian cities via the Tigris and its bypass channels and that links with the northern areas were well established. When trying to explain this preferential trade route, we should take the Ur III political situation into account: provinces and cities conquered by the king Shulgi were obliged to pay a tribute (Roux, 1992: 171). We know that several cities were famous because of their specialized productions. Tello was famous for its fish trade and perfume ointments, while Sippar was known for its paint production (Crawford, 1973: 233). Kimash was famous for its bitumen, a product abundant in that region, and was controlled by the kingdom of Ur. The capital thus must have imported its bitumen from Kimash since it was under its direct control. This solution, if our hypothesis is valid, was possibly the most profitable option for the city of Ur.

In addition it was still exclusively northern Iraqi bitumen which reached the Persian Gulf up to about 2200 BC. At this stage we must be precise that when we quote a northern Iraqi source as “Mosul area” we mean Mosul where we have several source data but also Kirkuk and Kifri which correspond to Nuzi-Kimash area as mentioned above. It is difficult to find an explanation for this: did only Assur deal with the settlements in the Persian Gulf? Or could it have to do with the quality of bitumen? Bitumen from the Hit area was very pure and therefore must have been highly appreciated, and perhaps Sumer preferred to keep this fine quality product for itself, and exported only second rate products. Sumer only exported lowest quality wool and textiles to the Persian Gulf in the second half of the third millennium BC (Potts, 1990a: 150), and it may be possible that this was also the case for bitumen. Screening of many samples from various archaeological sites in the Gulf though, has never identified any lumps of pure Hit bitumen. Lumps of pure Hit bitumen were found at Mari (Connan and Deschesne, 2007) but this feature is not surprising due to the relative proximity of this archaeological site to the Hit source. Most samples are archaeological bituminous mixtures with a vegetal and mineral input, and therefore are mixtures. These data strongly suggest that it was not pure bitumen that was exported to the Gulf but manufactured mixtures. The mixture preparation may have also entailed the mixture of bitumen from different sources which may partly obscured the geochemical properties allowing difficulties in identification of sources as underlined in the Akkaz study (Connan, 2011). These mixtures were prepared by specialized craftsmen, and then poured into jars to be exported as such. These jars were unloaded from boats and finally stored in warehouses where they have been discovered through recent excavations.

3.2.6 Bitumen in Iran

After having discussed the Uruk changes about bitumen trade in Mesopotamia, we should take a look at southwestern Iran where changes were happening as well. We must underline the fact that the present day knowledge in that part of the Near East is too limited to produce a reliable overview of changes through time. However, we intend to propose some preliminary ideas, drawn from the available data.

According to the results of the study conducted by Marschner *et al.* in 1978 on samples older than the Proto-Elamite period (3200-2700 BC), Farukhabad but surprisingly also Shafarabad were supplied by bitumen from the Ain Gir seepage, while several sites from the Susiana plain (Susa, Chogha Mish, Djaffarabad) imported their bitumen from Masjid-i-Suleiman. This conclusion appears reasonable taking into account the geographic locations of both archaeological sites and oil seeps. However these interpretations must be considered with much caution for they derived from the plot of color (six-point scale) vs. size (% finer than 74µ) of mineral particles remaining after the removal of bitumen. In other words, the conclusion about sources of bitumen does not rely on the bitumen itself but on the mineral fraction present with the bitumen in the sample. This fraction is not necessarily related to the oil phase and may have been added onsite. No molecular or isotopic data on the bitumen phase were produced in this pioneering study, published in 1978. If efficient tools had been available, the bitumen-to-oil seep correlations would have been easier to establish, for Masjid-i-Soleiman oil seeps do contain 18 (H)-oleanane in relation to the Pabdeh Tertiary source rock contribution whereas the Ain Gir oil shows to originate from a different source without 18 (H)-oleanane. Such investigation has been carried out more recently with bituminous mixtures from Susa, covering a large time scale from the Ubaid 4 (Period I-II of Susa, 4200-3100 BC) to the Parthian occupation (200 BC-200 AD). The results (Figure 12) shows that the import of bitumen with 18 (H)-oleanane is recorded all along the history of Susa but that several other sources without 18 (H)-oleanane (Ain Gir, Sultan) are also encountered. No real preferential utilization of one particular source has been noted when screening the available data, but the low density of information, especially during the earlier periods, may be responsible of this truncated pattern.

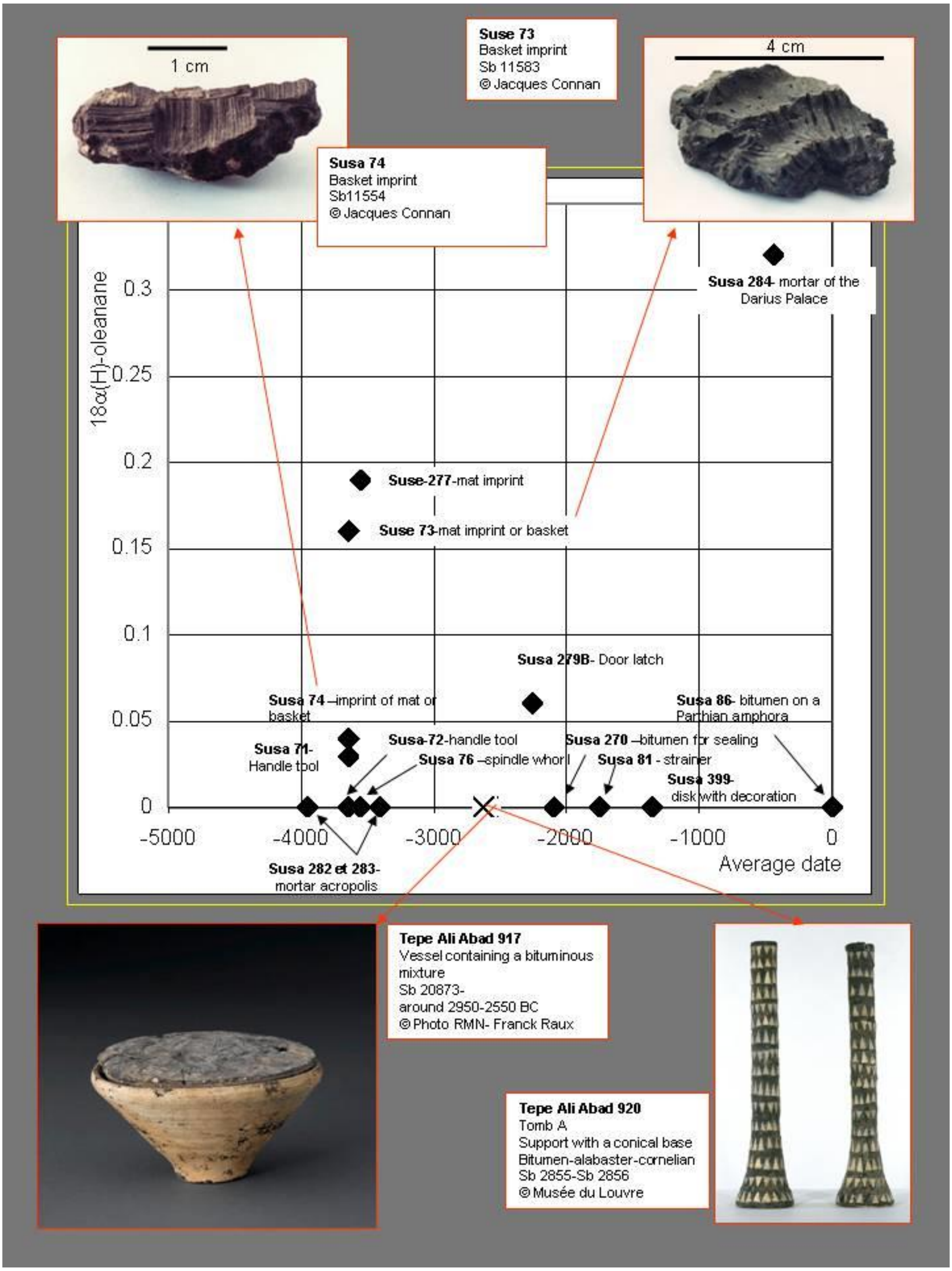

Figure 12 Distribution of 18α(H)-oleanane through time in various bituminous mixtures of Susa.

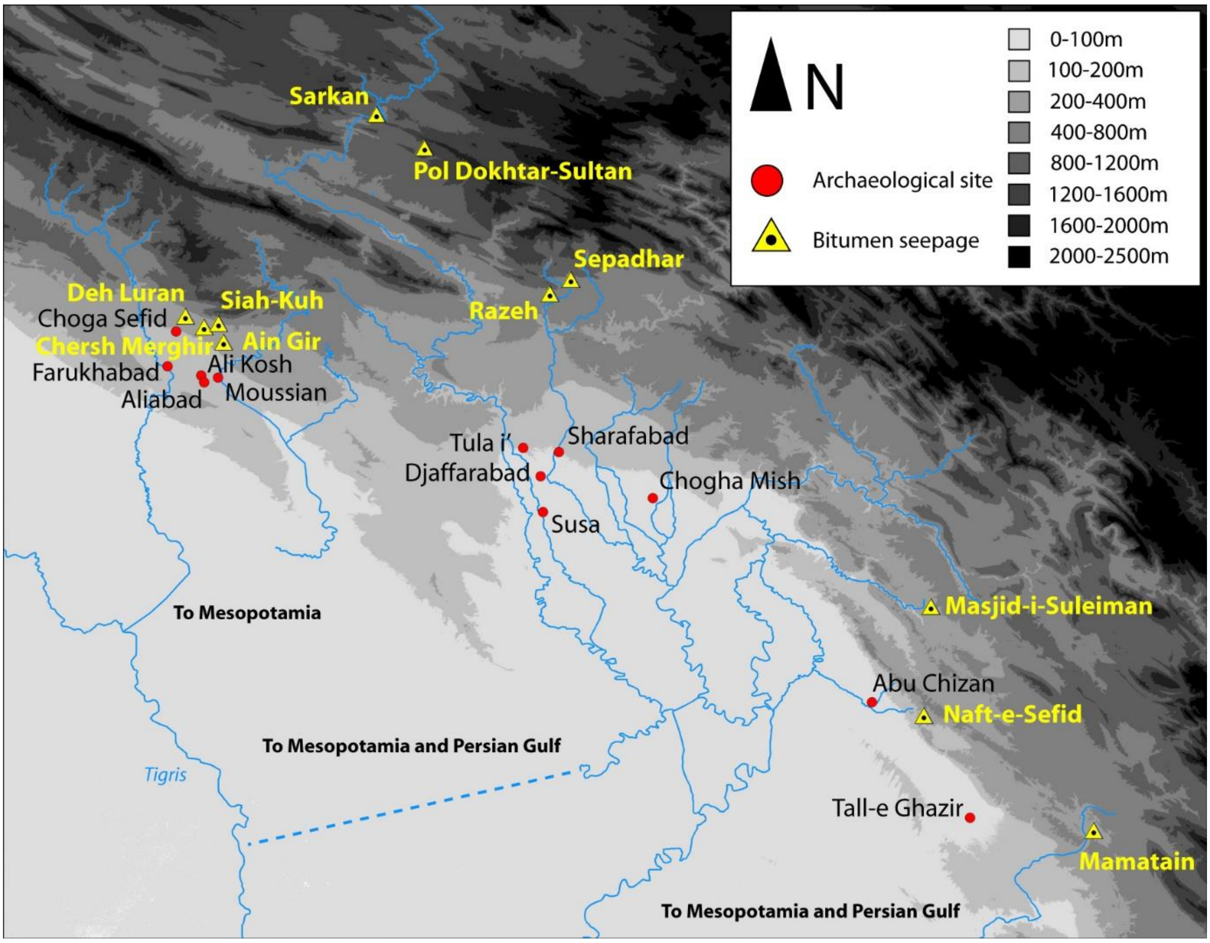

Figure 13 Overview of the Deh Luran and Susiana plains in southwest Iran. All well-known bitumen sources are located along the rivers, providing easy transport. It appears that each plain had their own bitumen sources and exchange of resources was not a necessity, but nonetheless attested from the Proto-Elamite period onwards.

However the proposed overview for the bitumen supply fits with what is seen with other products manufactured in this area: for example, it is known that during the Middle Uruk period, Chogha Mish and Susa had their own distinctive pottery style spread within the smaller settlements in their vicinity (Wright and Johnson, 1975: 280). At about 3200 BC, the start of the Proto-Elamite period, Marschner *et al.* (1978) proposed occurrences of bitumen from Masjid-i-Suleiman at Farukhabad and bitumen of Ain Gir at sites in the Susiana plain. These changes can be attributed to the changing political and social climate in southwest Iran at that time. In the second half of the fourth millennium, the different settlements and tribes in the Zagros mountain united and formed the Proto-Elamite civilization (3200 – 2700 BC). Thanks to the new unity, the economy prospered and there was a remarkable growth of population, leading to a higher need for raw materials and consequently an intensification of the trade network, possibly encouraged and/or sponsored by Mesopotamian Uruk merchants (Alden, 1982: 622). It is during this time that the concept of 'city' was introduced in southwest Iran as well, which brings along, exactly the same as in Mesopotamia, monumental architecture and an associated greater demand for raw materials. The changing of culture, urban life and socio-economic behaviour might explain the longer distance trade network of bitumen export from various sources during this period. This general exchange scheme seems to be confirmed by recent studies for an analysis of bitumen from Tall-e Ghazir reveals that its bitumen did not come from the neighbouring sources, either Naft Safid at 35 km or the Mamatain oil seepages at 30 km, but originated from another source without 18 (H)-oleanane, which is presently not fully identified but which shows some similarities with Sultan-Pol Dokhtar, located 250 km away (Connan et al., 2014). This bitumen has been found at Susa and may have been imported to Tall-e Ghazir via this city. Tall-e Ghazir is strategically located on the trade routes linking lowland Mesopotamia and Susiana to the highlands and points east, as reflected by its ceramics which combine both the highland and lowland traditions (Alizadeh, 2006) (Figure 13).

3.2.7 The Persian Gulf from 2200 B.C. onwards

We have already stated that prior to 2200 BC, bitumen from exclusively northern Iraq reached the Persian Gulf. In the late third millennium BC, trade in the Persian Gulf area bloomed and a dense maritime network was created. This made Mesopotamia materials and products more accessible for settlements on the Persian Gulf shores, and we see that from this period onwards bitumen became a more common product in the Gulf (Connan and Carter, 2007). From this period, bitumen was increasingly used, for instance in the architecture of Bahrain, as pointed out by Højlund and Hendersen (Højlund and Andersen, 1994). One must underline here that the oil seeps are very scarce along the southern coast of the Gulf and limited to two well-known oil occurrences, Burgan Hill in Kuwait and Djebel Dukkhan in Bahrain, to which the famous Haushi oil seep (Figure 14) in Oman has to be added. The latter, which is situated around 200 km from Abu Dhabi and 160 km from the Omani coast, have not yet been identified in samples from archaeological sites along the coasts of the Gulf and Oman. Oil seeps from the Djebel Dukhan, on Bahrain Island, have not been identified in any archaeological mixtures from sites located in the Gulf and Oman. Consequently the utilization of bitumen in the countries of the southern coastline of the Gulf was hitherto limited to the immediate vicinity of oil seepages, and was not a common practice at many other settlements. It is only following the expansion of the international maritime trade routes in the Gulf that the generalized utilization of bitumen took place. According to modern economic concepts, the Gulf offers a demonstrative example of a so-called technology transfer, made possible with adequate means of communication, especially seafaring boats.

The expansion of long-distance trade can be attributed to the changing copper routes. Copper, a necessary material for Mesopotamia, was now no longer imported from Iran, but from Oman (Potts, 1993c: 391). Due to this dense trading network, cities like Qala'at al-Bahrain and the settlements on Failaka Island (notably Tell F6) became very important players in long-distance trade, functioning as ports-of-trade. Several of these cities were successful in situating themselves within the new trade network, making them rich and prosperous. The city of Qala'at al-Bahrain for instance, saw an immense economic growth from the late third millennium onwards and succeeded in promoting itself as the most important city on the Bahrain Island (Killick and Moon, 2005b: 347).

To secure these trade routes, Mesopotamia became closely involved in these beneficially located harbour-cities in the Persian Gulf, as is clear from both textual and archaeological evidence; this is noticeable in the pottery and the Mesopotamian influence on the stamp seal iconography (Højlund and Hellmuth Andersen, 1997: 85). Remarkably the settlements under Mesopotamian influence used exclusively bitumen from Hit, while sites that do not share this special connection with Sumer imported their bitumen from other areas (north Iraq or Iran). This is clearly documented in the case history of Bahrain. After 2200 BC it is bitumen from Hit which is exclusively found at Qala'at al-Bahrain but other third and second millennium sites on the island, notably Karranah, Buri and Saar, imported their bitumen from Iran. The difference in bitumen supplier is not the only thing that differentiates these settlements from each other: the occupation levels at Saar, for instances show some Mesopotamian products and materials (Crawford, 19991: 17), while these were abundant at Qala'at al-Bahrain (Lombard, 2000, Lombard, 2004). This indicates the privileged bond that Qala'at al-Bahrain shared with Mesopotamia. Qala'at al-Bahrain may have profited from this partnership by importing bitumen at more profitable rates (Connan et al., 1998). When we take a closer look at Saar, we see that the inhabitants imported their bitumen from southwest Iran (possibly the Khuzistan area). This implies not only that the settlement had no strong bonds with Mesopotamia but also that there must have been Iranian merchants present in the island or Bahraini merchants in Iran. The occupation levels of the settlement had little Iranian material: only 2 beakers with a possible Iranian provenance (Carter, 2005a: 264) and one bronze sample undeniably had an Iranian origin while the bulk of bronze materials had a southeast Arabian provenance (Weeks and Collerson, 2005: 323) as would be expected regarding the bronze trade from Arabia to Mesopotamia. The bitumen, however is Iranian, which implies that its import may have been a specialized activity. Another possibility is that the inhabitants of Saar, Buri and Karranah did not want to associate themselves with the Mesopotamian dominion at Qala'at al-Bahrain and found themselves more '*Dilmunite*', and therefore used other commercial connections (Connan et al., 1998: 177). But again, this means that they must have been in contact with Iran through a trade network. More evidence of such a network of trade and influence was noted through the study of Kaftari- and Kaftari-related ceramic vessels. Though limited only to the late 3rd and early 2nd millennium BC, such vessels were identified at Failaka (F6), Bahrain (Qal'at al-Bahrain, Dar Kulayb), eastern Arabia (ar-Rafiah) and southern Arabia (Tell Abraq, UNAR 2) (Petrie et al., 2005). Unfortunately, we have no analyses from sites from the Oman Peninsula from the late third or early second millennium BC, but we can assume that, similar to Saar, they just imported their bitumen from the cheapest or easiest accessible supplier, possibly Elamite partners.

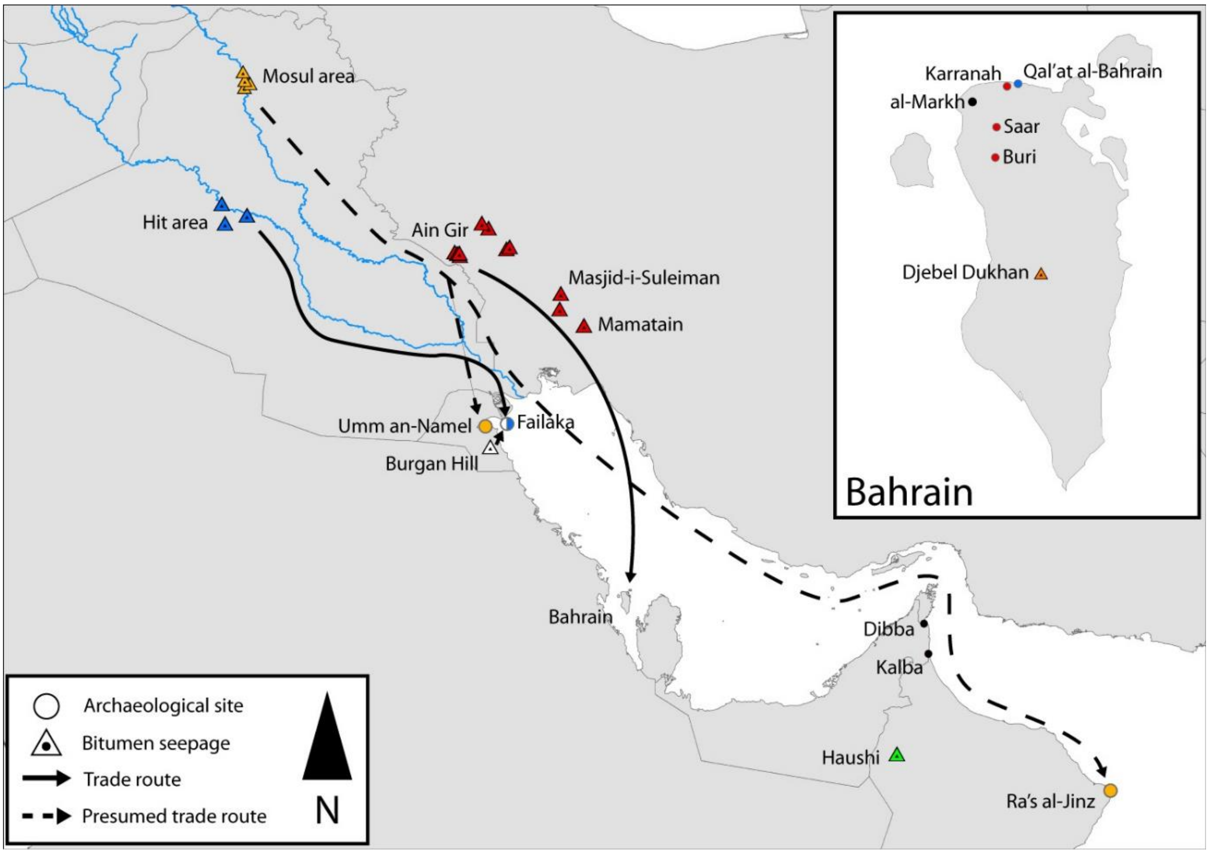

Figure 14 Bitumen export to the Persian Gulf from 2200 B.C. onwards. Failaka and Qala'at al-Bahrain could rely on a steady flow of bitumen import from Mesopotamia whereas other sites had to import their bitumen from elsewhere. At the present state of the research, there is no evidence of bitumen exploitation from Djebel Dukhan (Bahrain) nor from Haushi (Oman).

The import of exclusively Hit bitumen to Qala'at al-Bahrain came to an end at about the fifth century BC, the ending of the Late Dilmun period. This period also marks the downfall of the last Mesopotamian kingdom (Neo-Babylonian period), and this means that Qala'at al-Bahrain and Failaka lost their privileges and had to take care of their own import of raw materials. Both cities kept on importing from Hit, though not exclusively. We see that Qala'at al-Bahrain imported now not only from Hit, but from Iranian sources as well.

The appearing of Iranian bitumen is also attested at the archaeological sites of Akkaz and ed-Dur, both belonging to the early centuries of our era (Connan and Carter, 2007). At the site of ed-Dur, archaeologists unearthed bitumen originating from the Mosul area as well, up to the fourth century AD. We see that for the fourth century AD, Mosul is the only bitumen supplier for ed-Dur. There is only one sample analysed for this period though, and this sample cannot stand for the entire period. The appearance of raw materials from at least 2 areas of bitumen exploitation points out the cosmopolitan character of this site, located strategically at the entrance of the Gulf.

It is quite remarkable that the settlements of Failaka and Umm an-Namel kept on importing their bitumen from the Hit area, while bitumen with an Iranian provenance is found on the site of Akkaz. It is possible that in that period Failaka was not conquered by the Parthian Dynasty (Connan and Carter, 2007). But what about Umm an-Namel? The samples from Akkaz and Umm an-Namel have an overlap of a hundred years, and it does seem questionable that the Parthians conquered Akkaz (and controlled the trade), while they left Umm an-Namel, merely 5 km away, undisturbed. Differences may be also related to a shift in trading patterns between Parthian and Sasanian Period. Pottery suggests a Parthian dating for Umm an-Namel, and Sasanian for Akkaz (Carter, personal communication). It is also possible that, similar to what happened at Bahrain in the Early Dilmun period, we have 2 different populations, each one assimilating itself with another party, one being the new Iranian Dynasty, another being the old Mesopotamian bond. Another possibility is that politics and ethnicity did not really matter at all, and each settlement just bought their resources from the merchant who could offer them at the lowest rates.

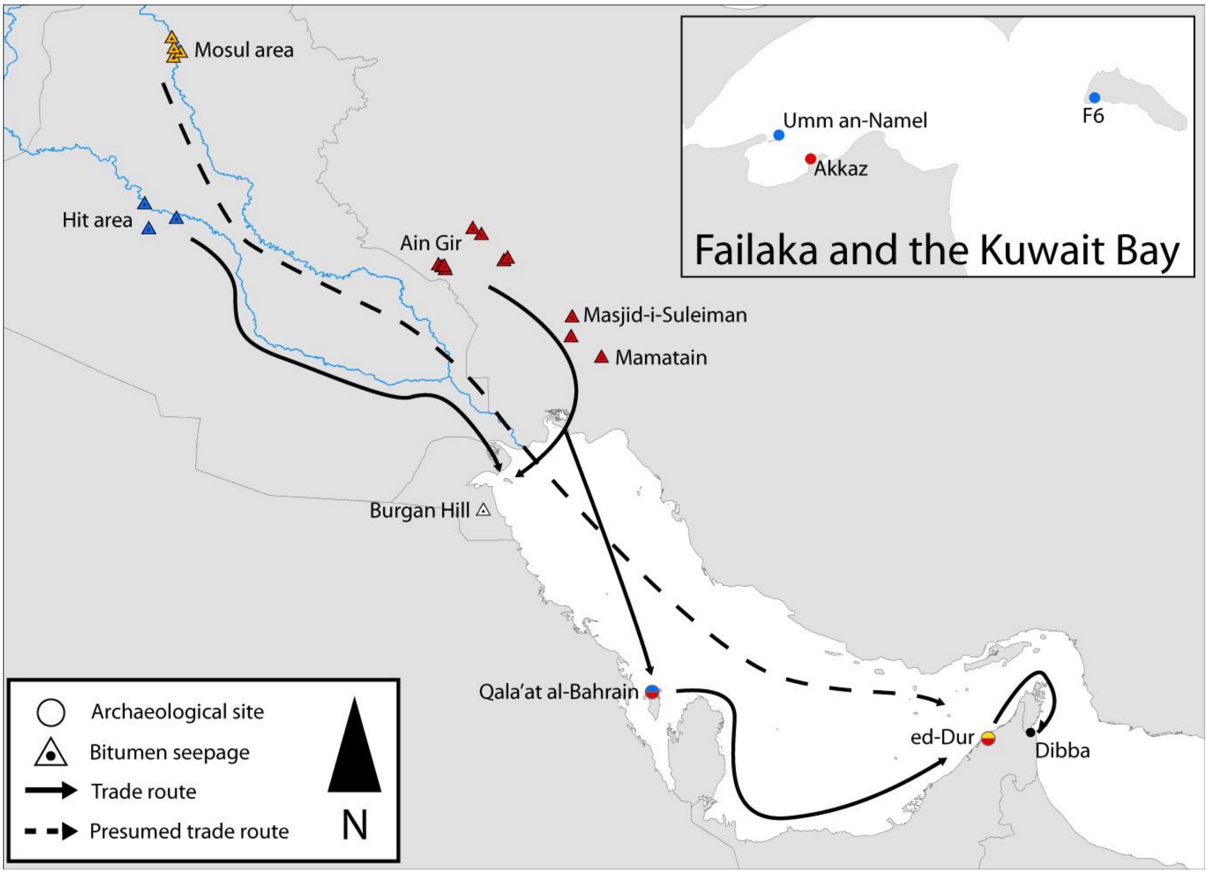

Figure 15 Bitumen trade routes in the Persian Gulf during the late first millennium BC.

It is noteworthy that Iranian bitumen seems to play a bigger role in the Gulf from the second half of the first millennium BC onwards, for this bitumen crossed the Gulf to supply the settlements in the SE-Arabia, and travelled all along the Oman coast and even across the Indian ocean as shown by its discovery at Anuradhapura in Sri Lanka (Stern et al., 2008). In Anuradhapura, however, the bitumen seemed mostly to have been used to seal or waterproof the jars, and consequently may have not been exported as mixtures in the analyzed potsherds to be sold and used locally.

As already noted, the available data suggest that bitumen was exported to sites in the Persian Gulf in the form of a mixture, and not as a raw material. Beside the absence of lumps of pure bitumen, this hypothesis is supported by the recent discovery of bitumen-filled jars of the 1ste century A.D. at Dibba (east coast of the U.A.E.), which were all mixtures.

3.2.8 Dead Sea bitumen

Though Mesopotamian and Iranian bitumens were widespread across the Near East, they are not found in the Eastern Mediterranean. Many sites in this area were provided with bitumen from the Dead Sea (Nissenbaum, 1978, Nissenbaum, 1994, Nissenbaum and Goldberg, 1980, Rullkötter and Nissenbaum, 1988, Spiro et al., 1983). Concerning this trade, three cities seem to have been of uttermost importance: Arad, Ein Besor, and Palmahim. These settlements were strategically located for the bitumen trade: Arad near the bitumen source, the coastal site of Palmahim for oversea transportation and Ein Besor which controlled "*The way of Horus*", an important overland route to Egypt (Connan et al., 1992: 2758). Via these trade routes, bitumen could be exported to Egypt or any coastal settlement in the Eastern Mediterranean. Proof that this was a profitable trade is found in bitumen analyses at Tell Miqne: in the second quarter of the twelve century BC, the Philistines (which are part of the so-called "Sea Peoples") arrived in the Eastern Mediterranean and caused an abrupt end to the Bronze Age Canaanite civilization who, after the invasion, were obliged to take refuge in the highlands of Judea and Israel, while the occupiers settled in the coastal areas (Connan et al., 2006b: 1770). Evidence from Tell Miqne points out that the new Philistinian settlements knew prosperity and economic growth, despite that they were constant at war with the Israeli tribes. It is remarkable that in this period of warfare, bitumen from the Dead Sea still reached Tell Miqne, which indicates that political and ethnical disagreements and warfare were subordinate to economic motives (Connan et al., 2006b: 1785). Recent analysis of samples from Yarmouth (de Miroschedji, 1999), showing mostly shiny lumps of bitumen with a conchoidal fracture indicating likely pristine asphalts, has indeed confirmed that all these archaeological samples were pure unprocessed Dead Sea bitumen (Connan, unpublished). Some samples that weighed 1 kg are mainly dated to between 2650 and 2200 BC but two samples were younger, between 1400 and 1100 BC. Dead Sea bitumen was exported to Egypt for funeral utilization (balms of human and animal mummies, coffins and statues paintings, funerary objects) and has been identified in many balms of Egyptian mummies (Rullkötter and Nissenbaum, 1988, Connan, 1999b, Connan, 2005, Harrell and Lewan, 2002, Maurer et al., 2002). The key difference with other famous bitumen from Iraq (Mosul and Hit) is that the Dead Sea bitumen is exported as pure lumps which may reach the weight of more than 1 kg. This statement oozes from archaeological finds (Edgar, 1905, Menghin and Amer, 1936, de Miroschedji, 1999) which document discoveries of dominant pure bitumen lumps.

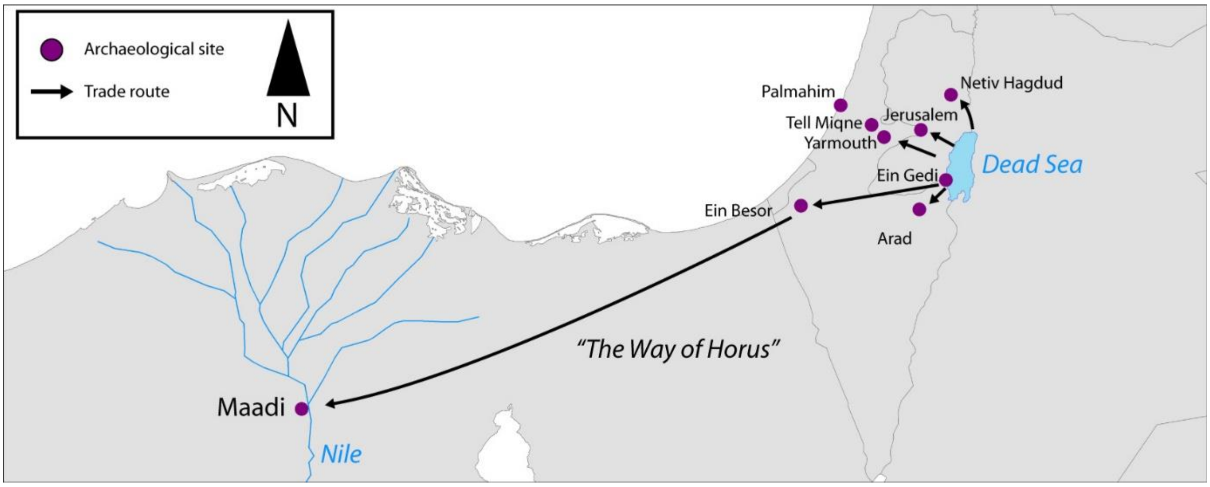

Figure 16 The export of Dead Sea bitumen.

3.2.9 Conclusions

As more and more data are acquired on new oil seepages or archaeological sites, more and more complexity appears in the bitumen trade of the Near East. Bitumen was used from the Palaeolithic or early Neolithic period when abundant raw material was available in the vicinity of certain settlements. When the source was actively producing bitumen in the neighbourhood, uses became diversified and frequent, as seen for example at Abu Chizan in Iran. When the sources were scarce and limited to small oil shows, some selected uses probably reserved for artefacts, devoted to an expected elite population such as beads or rings. In some cases even, its scarcity did not lead to a high valorisation. From the 6th millennium to the middle of the 4th millennium this situation was widespread due to the limited means of transportation, but the import of bitumen has been recorded in certain areas, where bitumen was exchanged along with other objects, cultural or goods.

Long distance trade in bitumen is evident from the Ubaid 0 period (Iranian bitumen which travelled to Tell'el Oueili) and the Ubaid 3 period (the presence of northern Iraqi bitumen in the Gulf), but this trade was rather limited. It is only in the Uruk period that an actual long distance commercial network was established, providing settlements with bitumen all along the river Euphrates. This is probably also linked to an increased demand for this raw material, for more prestigious buildings in newly developing cities. The bitumen trade network expanded in the Middle-Bronze Age (late-3rd millennium BC) when some settlements in the Persian Gulf received a steady flow of natural resources from Mesopotamia. As a consequence, sites along the southern coast of the Gulf, which were not at all familiar with the use of bitumen, change their behaviours and joined the bitumen-using civilizations by way of technology transfer. This habit was not abandoned throughout the last two millennia BC and continued until the Early Islamic period and beyond, as testified by occurrences during Medieval times. In the first millennium AD, the source of bitumen in the Gulf was diversified and the Iranian bitumen contribution was enhanced, especially under the Parthian and Sasanian dynasties. Within the bitumen story, Dead Sea bitumen had its own trade network with particular export routes in present day Israel and outside, towards Egypt for mummification purposes.

3.3 Some insights in bitumen

3.3.1 Chemical fractions in bitumen

Bitumen is a complex mixture of organic chemical compounds consisting of four main molecular fractions: Aromatics, Resins, Saturated Hydrocarbons & Asphaltenes (see Figure 17). Aromatics are chemical compounds consisting of one- or more benzene rings (aromatic hydrocarbons) whereas resins are solid or semi-solid mixtures of complex organic substances with significant nitrogen, sulphur and oxygen (Peters et al., 2005a: 357, 389). These two fractions do not hold any relevant information for identifying the source of archaeological bitumen. Asphaltenes are the heaviest components of petroleum fluids that are insoluble in light n-alkanes such as n-pentane or n-heptane, but soluble in aromatics (Goual, 2012: 28). Often, stable carbon isotope analysis ($\delta^{13}C$) is used on this fraction as the relation between the ^{12}C and ^{13}C isotopes of the asphaltenes is seepage-specific.

Saturated hydrocarbons are non-aromatic hydrocarbons in which the maximum number of hydrogen atoms are bonded to the carbon atoms (Reniers, 2008: 25). Gas Chromatography – Mass Spectrometry (GC-MS) is used on this fraction for the identification of biomarkers. These are particularly useful as their complex structures reveal a lot of information on their origins (Peters et al., 2005a: 3). Especially the latter technique has proved itself very useful in fingerprinting petroleum and is used commonly in the petrochemical sector. See Figure 17 for an overview of the fractions.

3.3.2 The development of an analytical toolset

The advent of identification techniques for petroleum was around 1967 when the supertanker SS Torrey Canyon struck on a stony reef on the southwest coast of the UK. The enormous oil spill caused by this accident led to the cooperation of the petroleum geochemistry, oceanography, geology, and environmental chemistry, resulting in pioneering work in the use of biomarkers to differentiate petroleum and background hydrocarbons (Peters et al., 2005a: 276) In such disasters it is important to perfectly identify the spilled oil; not just to identify the contaminator (if there is a dispute related to liability), but also to assess the natural resource damage in order to get an idea of the potential long-term impact on the environment and to select the appropriate measures for clean-up (Wang, 2008). Currently, the oil spill identification system is largely based on GC-FID and GC-MS techniques for which the only the latter technique provides data on the source specific marker compounds (Wang and Fingas, 2005: 1037, 1041). Therefore it is useless to do GC-FID analysis on archaeological samples if the purpose of the research is the identification of the origin of the bitumen. Similar to oil spill research, the process of sourcing archaeological bitumen is based on matching, i.e. does the data from a sample (be it archaeological or from an oil spill) match any known oil field/seepage?

As mentioned above, archaeological bitumen is commonly a mixture of pure bitumen and a temper, prior to actual bitumen fractionation and analysis the organic matter needs to be separated from the inorganic materials. Separation of the organic from the inorganic matter was already executed in the pioneering study on archaeological bitumen by Marschner, Duffy and Wright (1978: 106). Their analytical procedure, however, differs from the current standards as the analytical tools which are now widespread were not available at the time (Connan and Van de Velde, 2010: 10). A workflow for bitumen analysis has been tried and tested consequently by A. Nissenbaum and J. Connan and used repeatedly with success (Connan et al., 1992, Nissenbaum and Buckley, 2012, Nissenbaum et al., 1984, Rullkotter et al., 1985, Connan, 2012, Connan and Deschesne, 1996, Boeda et al., 1996, Brown et al., 2014, Connan, 1988, Connan et al., 2004). Generally, chloroform or dichloromethane is used to extract the organic matter (i.e. the bitumen) from the archaeological sample, after which the sample was separated into different fractions. Isolation of the asphaltenes is achieved by precipitation by hexane, whereas HPLC or MPLC (High- and Medium Pressure Liquid chromatography) (Peters et al., 2005a: 199, Connan et al., 2004, Connan and Nissenbaum, 2004, Churley et al., Connan, 2012) is used for eluting the saturates. It is, however, not uncustomary to use standard open column chromatography in order to isolate the Saturated Hydrocarbons prior to GC-MS analysis (Connan et al., 2006b, Schwartz and Hollander, 2008, Stein et al., 1999). Generally n-hexane is used to elute the fractions in standard column chromatography, but also n-pentane can be used (Fuhr et al., 2005, Wallace et al., 1987).

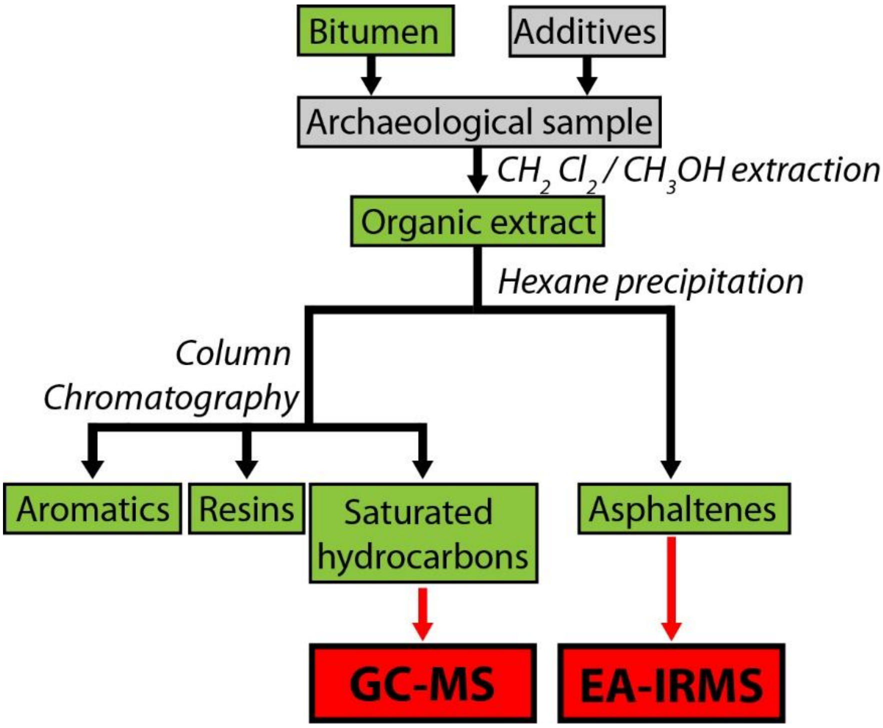

Figure 17 Overview of the fractions in bitumen including the methods of de-fraction and analytical techniques used.

Primarily only GC-MS on the Saturates was chosen to identify the origin of the archaeological bitumen samples. Although this method using biomarkers reveals the most information concerning the geological origin of the samples, it was later acknowledged that stable carbon isotope analysis on the asphaltenes would give valuable and additional information and resolve some issues that were experienced in several samples. Consequently, not all samples have undergone the latter technique and generally GC-MS analysis of samples was conducted prior to $\delta^{13}C$, meaning that sample preparation was executed twice in different laboratories.

3.4 Chemical Methodology

3.4.1 Sample preparation

Based upon literature and extensive try-out's, a methodology was chosen and followed for all of the samples analysed in Ghent during the course of the research. Generally, the aim was to use 1,5g of sample for analysis. This was often not possible and also much smaller samples (with a weight as low as 0,5g) have been successfully analysed. First, the archaeological sample was crushed for solvent extraction using CH_2Cl_2:Methanol (2:1 ratio). Generally the sample was prepared manually by treating the samples ultrasonically (20 to 30') and centrifuging (10' at 2000 rpm), after which the liquid fraction was recovered. This step was repeated twice in order to isolate as much of the bitumen as possible. The first step in the sample prep prior to stable carbon isotope analysis is the same as described above, but here an Accelerated Solvent Extractor (Dionex ASE 350) was used to automate the separation of the organic from the mineral matter.

For asphaltenes isolation, the obtained solution was dried using a rotovapor and then taken back into solution in hexane. The residue is the asphaltene fraction. This operation was repeated several times to be sure that no coprecipitated molecules were trapped in the asphaltenes matrix.

Standard column chromatography was used for separation of fractions prior to GC-MS analysis, as described by Peters et. al. (2005a: 200). A column was packed with silica and pentane:CH_2Cl_2 (3:1) was used as solvent. The eluted fraction, the so-called saturates, is recovered and ready for GC-MS analysis.

3.4.2 Instrumental parameters

For the GC-MS analysis a Hewlett-Packard 6890-5973 GC-MS system equipped with an Agilent Technologies HP-5MS column (30m x 0.25mm ID, 0.25µm) was used. Helium was used as a carrier gas, with a gas flow of 1.5 ml/min. One microliter of the saturated hydrocarbon fraction dissolved in CH_2Cl_2 was injected (in splitless mode). The oven temperature increased step-wise from 40°C to 250°C at 6°C per minute, and from 250°C to 300°C at 2°C per minute. The temperature was then held at 300°C for 30 minutes.

The carbon isotopic composition of the asphaltenes fraction was determined using an elemental analyzer (ANCA-SL, PDZ Europa, UK), coupled to a isotope ratio mass spectroscopy (IRMS) (20-20, SerCon, UK). The isotopic composition of natural samples (i.e. not synthetic isotopic enrichment) is reported relative to an international reference, using the so called 'δ' scale and is typically expressed in ‰.

$$\delta^{13}C = \frac{\left[\frac{^{13}C}{^{12}C}\right]_{sample} - \left[\frac{^{13}C}{^{12}C}\right]_{VPDB}}{\left[\frac{^{13}C}{^{12}C}\right]_{VPDB}}$$

For C the international reference is Vienna Pee Dee Belemnite (VPDB), which has a carbon isotopic ratio $\frac{^{13}C}{^{12}C}$ of 0.0111802 (± 0.0000028).

For both techniques several replicate analyses of samples were conducted to cross-check consistency of the measurements. Generally no discrepancies were identified. If a sample was measured more than once, the average of all measurements was calculated and used for further analyses and interpretation.

3.4.3 Post-analysis processing

The outcome of stable carbon isotope analysis gives a value representing the relation between the isotopes ^{12}C and ^{13}C and is a single value for every sample. The $\delta^{13}C$ of a sample is seepage-specific and may consequently be used to determine the origin of the sample (see for Figure 18 $\delta^{13}C$ of the Iranian seepages and Figure 25).

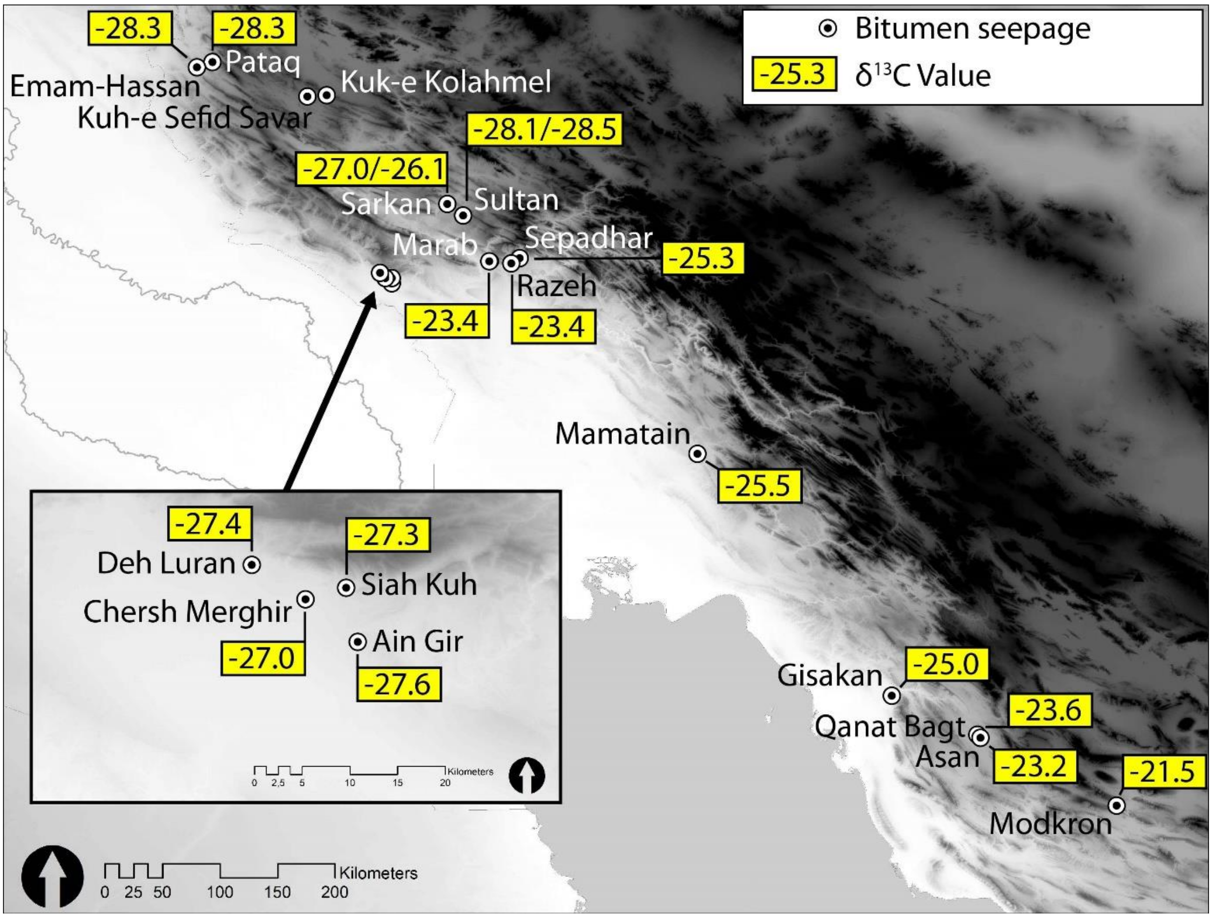

Figure 18 Map of Iranian bitumen seepages with their $\delta^{13}C$ values. Modified after Connan 2012.

The result of GC-MS analysis on the other hand is a chromatogram which needs further processing prior to data extraction (see Figure 19). For bitumen fingerprinting, the sterane (m/z 217) and terpane (m/z 191) fingerprint deliver the most reliable information for biomarker identification and quantification. Agilent's software package MSD Chemstation Enhanced Data Analysis was used for peak recognition and total peak surface was used for compound quantification for the terpanes. Retention times were used for manual peak identification, based upon what was known from literature. Several standards were acquired from Campro Scientific to aid in this process. The following molecules may aid in the identification process (steranes):

- 18α(H)-22,29,30-trisnorneohopane (C_{27}) (Ts)
- 17α(H)-22,29,30-trisnorhopane (C_{27}) (Tm)
- 18α-oleanane ($C_{30}H_{52}$)
- 17α,21β-hopane (C_{30})
- 22S-17α(H),21β(H)-Homohopane (C_{31}) (22S)
- 22R-17α(H),21β(H)-Homohopane (C_{31}) (22R)
- Gammacerane ($C_{30}H_{52}$)

Generally all of these compounds are present in archaeological bitumen (with the exception of oleanane) and may be used for fingerprinting, but there are several issues to be aware of. Not all of the molecules are equally stable and several of them are more prone to biodegradation than others. Also, individual compound intensities cannot be used for bitumen-to-oil seep (or bitumen-to-bitumen) identification; compound intensity ratios on the other hand provide a reliable tool for identification of archaeological samples. The most reliable compound ratios are Ts to Tm and Gammacerane to C_{30}αβ-hopane. Beside these ratios, the presence or absence of oleanane also gives information on the source of the bitumen, as this marker of both source input and geologic age is only present in Cretaceous and older oils (Peters et al., 2005b: 572). For the Near East and the area which is at study here, this is limited to oils originating from the Padbeh source-rock formation in southwest Iran (Connan, 2012: 117). Figure 20 shows the oil seeps in which chemical oleanane is present.

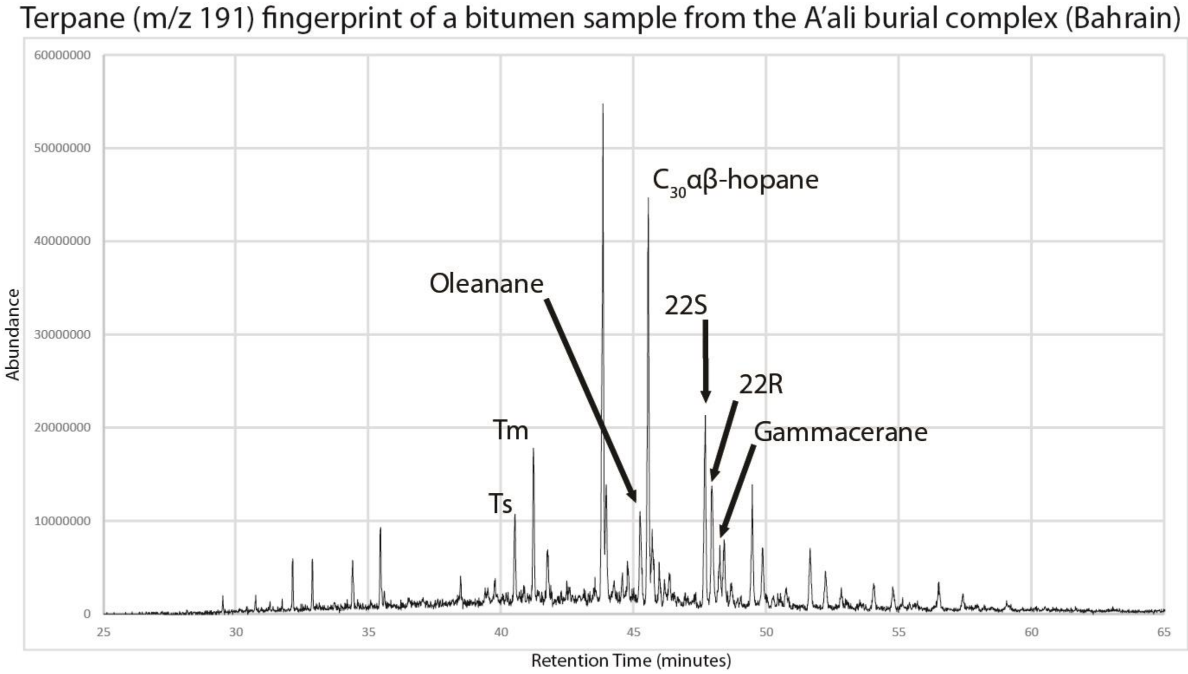

Figure 19 Terpane fingerprint of an archaeological sample coming from one of the Royal Mounds of the A'ali Burial Complex on Bahrain (Bronze Age).

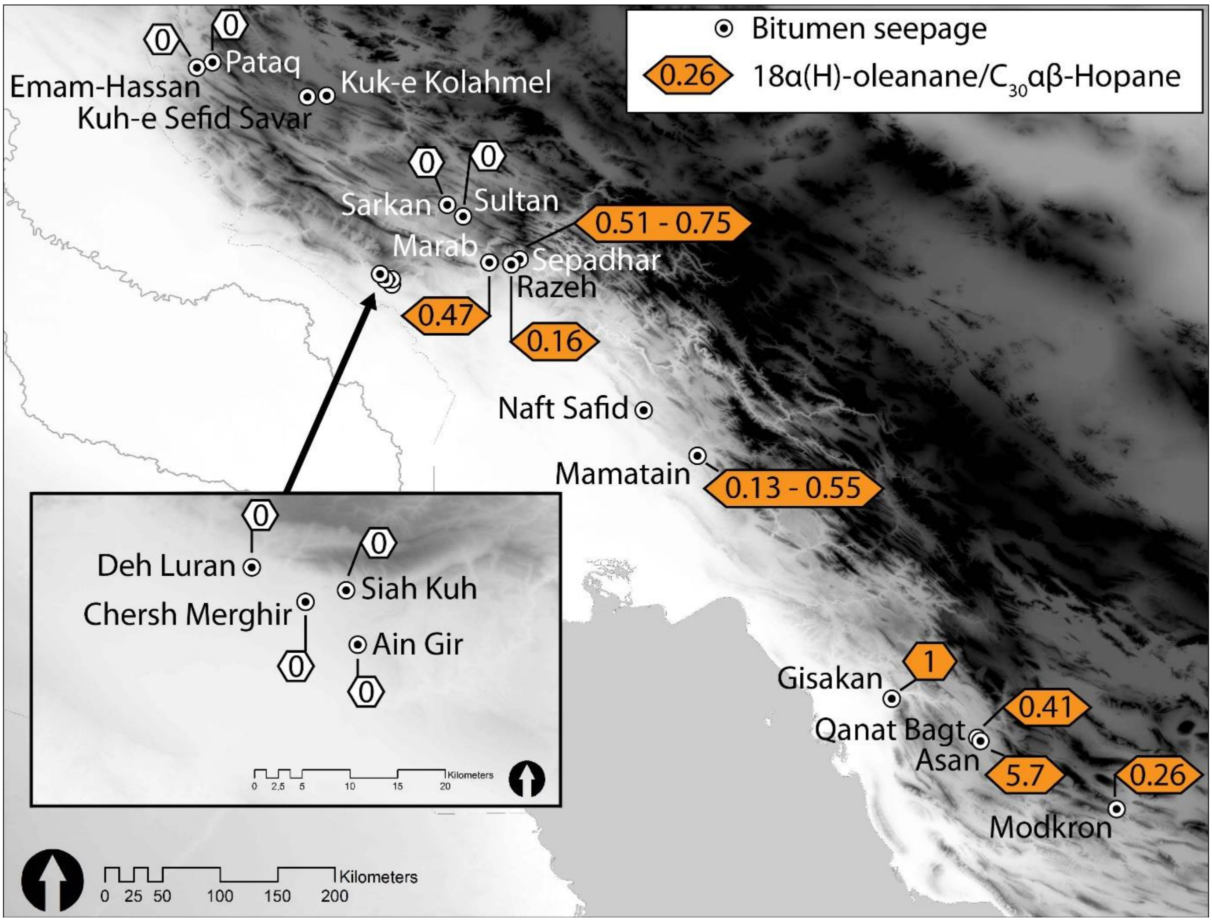

Figure 20 Iranian bitumen seepages and their oleanane/C_{30}αβ-Hopane values. Seepages with the value 0 do not contain any oleanane.

3.4.4 References

As highlighted above (See Chapter 3.3.2), the technique of fingerprinting archaeological bitumen relies upon matching a sample of unknown origin (i.e. the archaeological sample) with bitumen from a known origin (i.e. bitumen from an analysed seepage). This means that a positive match can only be made when reference data from the source origin is available, and exactly herein lie the challenges. Bitumen seepages that may be active today, were not necessarily so in Antiquity, and vice versa. This poses a problem in the research since the only way to tell whether or not a present-day seepage was active in the past, is by identifying it's fingerprint in the archaeological material. It is also very well possible that seepages disappear, due to tectonic activities for example. And archaeological bitumen to oil seep correlations are limited by the set of references for which geochemical data is available. Unfortunately this data is scattered and a complete database with all chemical properties of oil seeps in Iran (for example) is yet to be achieved (Connan et al., 1996). On a more commercial level, databases specific for the petroleum industry exist (with GeoMark as one of its main contributors), but were for this research unavailable due to the extremely high costs. Such a database would also not contain all the information necessary for sourcing archaeological bitumen. This is because petroleum from surface deposits may have a different chemical fingerprint than its counterpart from the reservoir, as the material weathers in its way to the surface, or even mixes with other oil. So a database with all the chemical characteristics of the current oil fields would not contain the same information as one with information on the reservoirs, and would therefore not fit the application here at hand. There are however quite some (academic) publications on the geology of petroleum systems in the Near East providing very useful information. Besides that, research has also shown that the match in molecular ratios is better when considering bitumen from archaeological sites rather oil seeps as molecular changes occur when working source material into bitumen mixtures (Connan and Carter, 2007: 65). Therefore, mainly archaeological bitumen datasets were used for the fingerprinting of the samples, many of which have been published in detail in many scientific journals and publications. The research presented in this manuscript embodies several bitumen datasets all from different archaeological sites and contexts, most of which have already been published or are currently in publication. All these scientific contributions are bundled here as different chapters in this book (see Chapters 4 to 6). The used bitumen reference datasets are presented separately for every chapter as these differ depending on the initial finding and the obtained results per dataset.

3.4.5 Processing of the data

Both molecular ratios and isotopic values were used to fingerprint archaeological bitumen when possible; unfortunately it was not possible to conduct stable carbon isotope analysis on all archaeological samples.

In order to successfully determine the origin of archaeological samples, they have to be compared with reference samples. It is practically impossible to use all of the available references for every archaeological samples, and reference datasets are chosen based on the initial findings of the archaeological datasets. In the first place, very source-specific items are considered. For example, bitumen with a $\delta^{13}C$ lower than -28.00 are limited to Hit bitumen (in regard to Near Eastern seepages), and samples showing a clear presence of the oleanane-compound are limited to seepages related to the Padbeh source-rock formation (southwest Iran). And samples with a Ts/Tm compound ratio lower than 0,2 are more often than not coming from the Dokthar-Sultan seepages (Iran, see Figure 20). In this regard, reference datasets are selected. The archaeological- and reference datasets are then used as input in the IBM SPSS Statistics software package. Primary, the data is visually plotted and investigated. Very often, clear groups can be observed, but more often the clustering is unclear. This has obviously to do with the organic nature of the material and the variations that may occur within the sample. Therefore, statistical analysis is used to identify reliable clusters. The method generally used for this is Furthest Neighbour and (Squared) Euclidean as a measure.

Part 2 - Geochemical analyses on bituminous materials and its interpretations

Chapter 4 Bitumen in the Arabian Neolithic

4.1 A geochemical study on the bitumen from Dosariyah (Saudi-Arabia): tracking Neolithic-Period bitumen in the Persian Gulf

The content of this chapter has been published in:
Journal of Archaeological Science,

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Abstract

This chapter presents the detailed results of a series of geochemical analysis conducted on 20 bitumen samples from the Neolithic site of Dosariyah (Eastern Province, Saudi Arabia). The aim of this study was to establish the geological origin of this bitumen in order to identify the bitumen seepage they were extracted from. The majority of the samples could be successfully related to bitumen seepages of northern Iraq. Two samples didn’t match the bulk of the samples and probably came from the Burgan Hill seepage (Kuwait). Three samples were too badly degraded in order to deliver reliable data, and were removed from the dataset. These conclusions stand in contrast with H3/as-Sabiyah, —a site culturally and geographically linked with Dosariyah— which bitumen came exclusively from the Burgan Hill. In this paper we argue that this difference may be explained by the difference in dating between the two sites, as H3/as-Sabiyah slightly predates Dosariyah.

4.1.1 Introduction

Bitumen is the result of the thermal degradation of organic material in a sediment. Unlike oil, bitumen is indigenous to the rock in which it was found (i.e. it has not migrated) (Peters et al., 2005a: 360). The possibility however exists that the bitumen found a way through the sediment to the surface, forming a bitumen seepage. This natural phenomenon has been known for a very long time, and there is proof of exploitation of bitumen from these sources as early as the Middle Palaeolithic (Boeda et al., 1996). Analytical geochemical techniques —originally developed for the petroleum industry— were first used in the seventies to identify- and source bitumen from archaeological sites (Marschner et al., 1978). Following this pioneering work, numerous analyses have been conducted on archaeological samples from sites throughout the Near East. The aim of this article is to present the results of bitumen analysis on a sample set from Dosariyah.

A wide sphere of cultural interaction along the shores of the Persian Gulf during the 5th millennium BCE is known since the initial discovery of Ubaid pottery at the site of Dosariyah, Eastern Province, Saudi Arabia, in 1968 (Bibby, 1970, Burkholder, 1972). During subsequent years, similar pottery has been found along the southern coast of the Gulf between Ras al Khaimah in the lower Gulf and H3/as-Sabiyah and Bahra in Kuwait (Beech et al., 2000, Beech et al., 2005, Mery and Charpentier, 2013, Burkholder, 1984, Drechsler, 2011, Carter and Crawford, 2010, Uerpmann and Uerpmann, 1996, Boucharlat et al., 1991, Phillips, 2002, Vogt, 1994, Masry, 1974). Geochemical analyses carried out on a selection of Ubaid sherds proved their origin in southern Mesopotamia (Oates et al., 1977, Roaf and Galbraith, 1994). The study of bitumen sheds new light on the cultural contacts that the inhabitants of Dosariyah had with their neighbouring cultures.

4.1.2 Materials and Methods

4.1.2.1 Archaeological samples

Archaeological field work at the Middle Neolithic site of Dosariyah, dating into the first half of the 5th millennium BCE, documented not only more than 9000 pieces of Ubaid pottery, but also registered a total of 244 bitumen objects and -lumps. In six of the eight trenches excavated by the *Dosariyah Archaeological Research Project* (DARP) bitumen was found (see Figure 21, Figure 22, Figure 23, Figure 24). When bitumen present, it was documented all across the stratigraphic sequences with the exception of the uppermost parts. Most plausibly, taphonomic processes led to the disintegration of bitumen in the upper layers, leaving behind a brownish sediment colour that is characteristic for many sediment layers at the site.

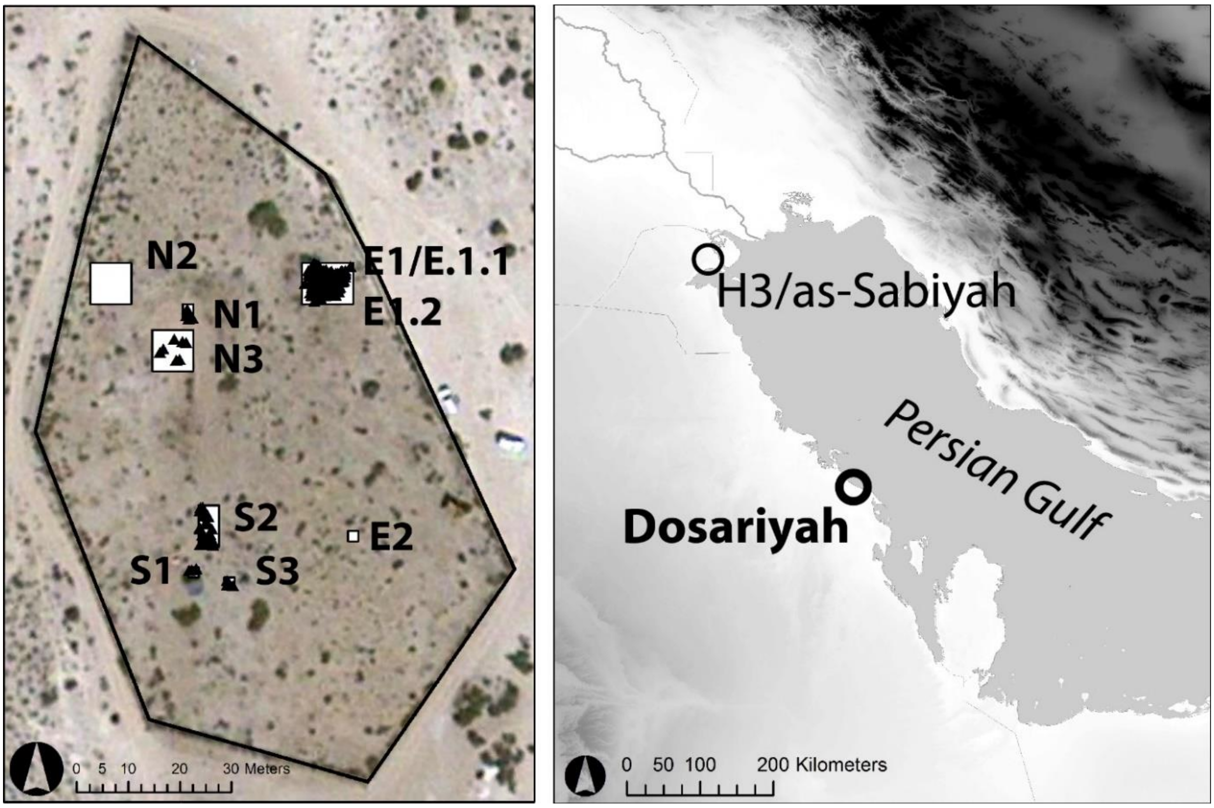

Figure 21 Localization of the archaeological sites of Dosariyah and H3/as-Sabiyah (right), and the site of Dosariyah with trenches and bitumen find spots marked (left).

Bitumen was not exclusively found as (part of-) artefacts, but mainly as shapeless lumps and fragments (as is common). From all registered bitumen pieces, a total of 20 were selected for analyses that derive from different archaeological contexts (all lumps, considering the destructive nature of the techniques used). The goal of these analyses was to establish the geological origin of these samples by using geochemical techniques which haven proven successful in the past (Connan, 2012, Connan, 1999b). These techniques embody essentially GC-MS analysis on the Saturated Hydrocarbon fractions and Stable Isotope Analysis on the Asphaltenes fraction (cf. infra).

The twenty selected samples were excavated in three different trenches and came from several sediment layers spanning the major part of the stratigraphic sequence. Most of the bitumen selected for analysis came from trench E1.1 (n=15), an excavation unit which yielded beside a complex stratigraphy also numerous features (fireplaces, pits, post-holes, and an unidentified pan-shaped structure with annex) and a high density of finds. Bitumen was found in almost all stratigraphic units of the trench (with the exception of the two uppermost horizons), although unit E1.1-III contained considerably more artefacts and marked a horizon of intense occupation.

Table 2 The samples analysed in Ghent with correlation to the Dosariyah trenches and object numbers. The remark ‘results not included’ means that these samples were not further processed due to low-quality chromatograms, which would lead to unusable data.

DOS sample n°	Year	Trench	Remarks
2613	2010	S1	Results not included
14330	2011	S2	
14668	2011	S2	
16190	2011	S2	
19554	2011	S2	
25364	2012	E1.1/E1.2	
26038	2012	E1.1/E1.2	
26341	2012	E1.1/E1.2	
26531	2012	E1.1/E1.2	
26551	2012	E1.1/E1.2	
26607	2012	E1.1/E1.2	
27083	2012	E1.1/E1.2	
28266	2012	E1.1/E1.2	
28344	2012	E1.1/E1.2	Results not included
28814	2012	E1.1/E1.2	
28912	2012	E1.1/E1.2	Results not included
29197	2012	E1.1/E1.2	
29239	2012	E1.1/E1.2	Crushed shell inclusions
30852	2012	E1.1/E1.2	
30994	2012	E1.1/E1.2	

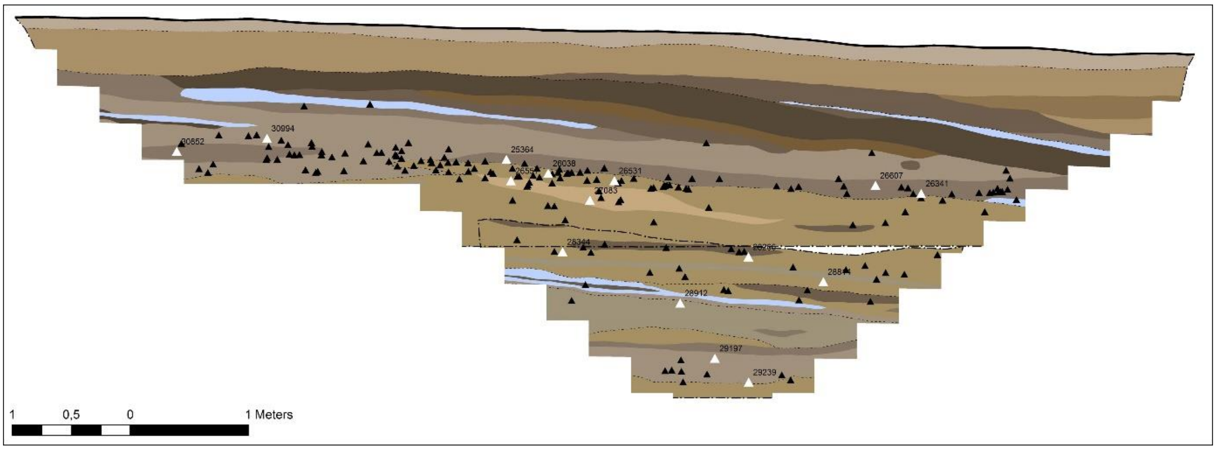

Figure 22 North section of trench E1.1. All bitumen finds are marked with triangles. White triangles symbolize bitumen pieces which were selected for analysis.

Trench S1 is a small test trench with shell-dumping accumulations in the upper part of the stratigraphic sequence, and with occupation horizons in the lower part of the trench. Of the four bitumen artefacts found here, one was used for detailed geochemical analysis.

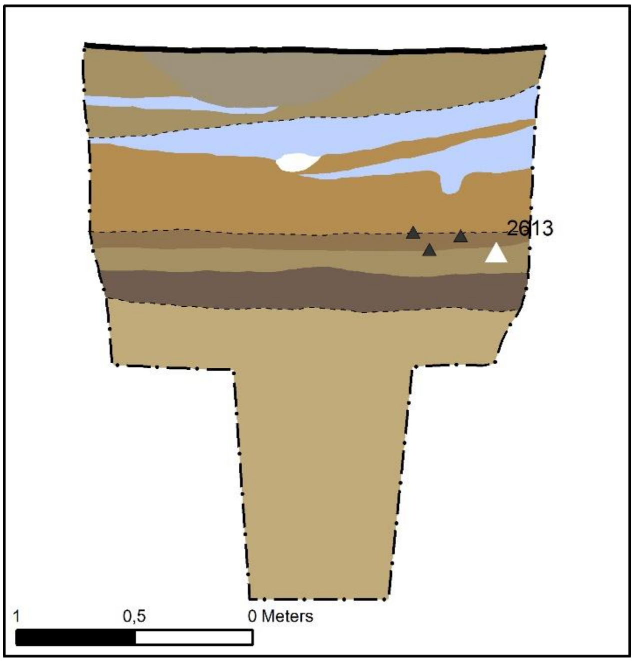

Figure 23 West section of trench S1. All bitumen finds are marked with triangles according to the respective layers in which they were found. White triangles symbolize bitumen pieces which were selected for analysis.

Finally, four bitumen pieces from trench S2 were sent to Ghent for analysis. The main stratigraphic features in this trench are similar as those from trench S1 (i.e. a shell accumulation in the upper part of the sequence, and occupation layers below), with the exception that there are pits and possibly postholes attested in this trench. Thirty-one bitumen pieces have been sampled from this trench of which four were subjected to analysis.

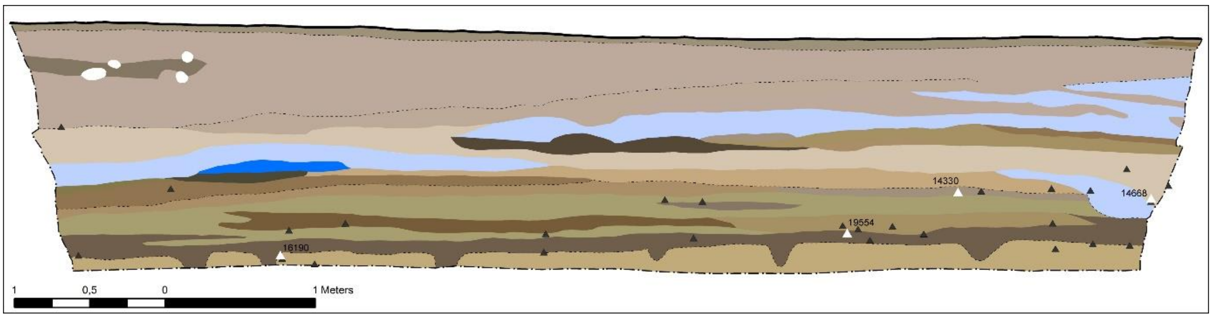

Figure 24 East section (x=1001) of trench S2. All bitumen finds are marked according to the respective layers in which they were found. White triangles symbolize bitumen pieces which were selected for analysis.

Although only a small fraction of all documented bitumen has been analysed (8.2%), the dataset mirrors the total number of bitumen found as the collected samples came from all parts of the site and various archaeological contexts and –layers.

Due to the destructive nature of the analyses, all selected objects were small shapeless lumps rather than identifiable artefacts. The weight of all samples varied between 0.6 and 2.5 grams. None of the bitumen from the selection was pure, but a mixture of different components (mostly a mineral matter such as sand), as is usual for bitumen artefacts (Connan, 2012, Connan and Deschesne, 1996). In the case of Dosariyah the bitumen objects were all very coarse with sand and tiny pebbles as visible inclusions. One sample (sample 29239) also had crushed shell inclusions, which is not an uncommon feature in bitumen samples. Considering that people in antiquity probably weren't too selective on the type of materials they used for bitumen mixtures, and the location of Dosariyah on the eastern littoral of the Arabian Peninsula, it's not surprising to see crushed shell used in bitumen mixtures. Several samples also showed signs of imprints, but always too small to identify any structures or shape.

4.1.2.2 Sourcing bitumen

Bitumen consists of several complex mixtures of molecules, some of which hold information concerning the geological origin. For sourcing archaeological samples, two different techniques exist, each developed for a specific chemical fraction (see Figure 17). The first fraction containing information on the source of the bitumen are the asphaltenes. These can be defined as the heaviest components of petroleum fluids that are insoluble in light n-alkanes (e.g. hexane) but soluble in organic compounds (e.g. dichloromethane) (Goual, 2012). Carbon Isotope Analysis (expressed as $\delta^{13}C$ of this fraction reveals information on the geological origin of the sample.

The second technique makes use of biomarkers. These are in essence molecular fossils; complex organic compounds derived from the original organic material the bitumen is formed of, which have a certain specificity for geologic origin. These biomarker are generally analysed using gas chromatography / mass spectrometry (Peters et al., 2005a: 3, 359). The terpane (m/z 191) and sterane (m/z 217) fingerprints give the most valuable information concerning the source of the samples.

4.1.2.3 Methods and references

The analytical methods as explained in chapter 3.4 were used for this dataset. Molecules present in the m/z 191 fingerprint were identified and quantified. Consequently, several source-specific molecule ratios were studied and compared with similar data found in literature. This other data can be original source data from actual bitumen seepages, or data from other archaeological bitumen. It should be noted that the match in molecular ratios is better when considering bitumen from archaeological sites rather than oil seeps as molecular changes occur when working source material into bitumen mixtures (Connan and Carter, 2007: 65). Molecular ratios from source-specific compounds from many archaeological sites have been published in detail by J. Connan in the past and provided a useful database for this research. The main references used for this research are the bitumen datasets from Kosak Shamali, Oueili, H3/as-Sabiyah and Ra's al-Jinz (Connan, 2010, Connan et al., 1996, Connan and Nishiaki, 2003, Connan et al., 2005). The same publications also hold data from $\delta^{13}C$ measurements on the asphaltene fractions. These datasets were chosen because of several reasons: Kosak Shamali, Oueili, and H3/as-Sabiyah were selected in the first place because they are (roughly) contemporary with Dosariyah. In the case of both Kosak Shamali and Oueili there appear to have been several bitumen suppliers which all could be candidates for Dosariyah. Ra's al-Jinz is a younger site (2nd half 3rd millennium), but its bitumen has been extensively published and therefore forms a reference framework for comparison purposes. See Table 3 for all used values in this chapter.

4.1.3 Analytical results

4.1.3.1 Determination of bitumen origin using Stable Carbon Isotope Analysis

Due to the limited size of samples it was possible to conduct this type of analysis on three samples only. As visible on Figure 25, the measurements indicate 2 groups. Two samples cluster together and fall within the range of bitumen from northern Iraq, as well as within a range of Ubaid 3-period samples from Kosak Shamali, Oueili, and samples from Ra's al-Jinz; all of which have effectively been identified as coming from northern Iraqi seepages (Connan et al., 1996, Connan and Nishiaki, 2003, Connan et al., 2005). The third sample (sample 27083) shows a significantly higher $\delta^{13}C$ (-27.2 ‰) and could correlate with reference values of seepages from northern Iraq, Iran, and the Burgan Hill. Data obtained from GC-MS analysis on this sample, however, favours a northern Iraqi origin for this sample (cf. infra). For measured values of $\delta^{13}C$ see Table 3.

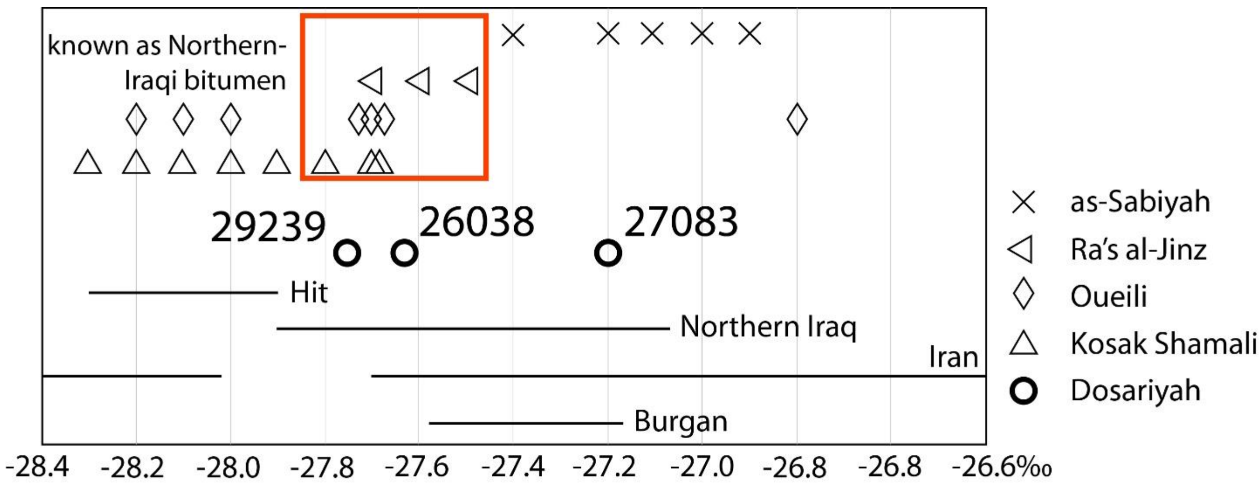

Figure 25 Graphic representation of the measured $\delta^{13}C$ (expressed in ‰) values of bitumen from various archaeological sites, and the ranges in which the source areas fall. The number represent the Dosariyah Find ID's.

4.1.3.2 Determination of bitumen origins using biomarkers

The terpane fingerprint (m/z 191) of all the samples is reminiscent to many other archaeological samples from the area with a domination of αβ-hopanes (see Figure 26). The lack of any 18α-oleanane in any of the samples rules out any origin in the Khuzestan- or Fars province in Iran. The main molecules used for fingerprinting this bitumen are 18α-22,29,30-trisnorneohopane (Ts), 17α-22,29,30-trisnorhopane (Tm), 17α,21β-hopane (C_{30}), C_{31}22R hopane (31R) and Gammacerane (GCRN). As the nature of organic materials such as archaeological bitumen mixtures does not allow a purely quantitative approach, ratios between individual molecules are used as source parameters.

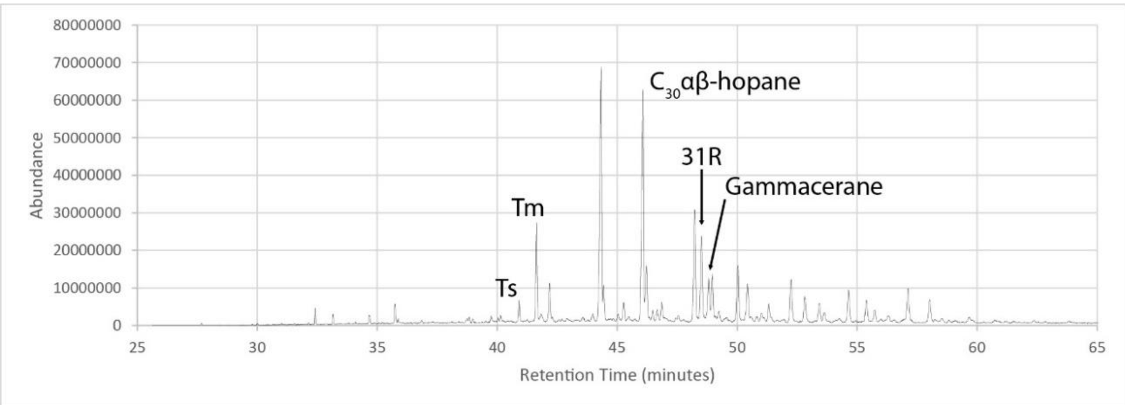

Figure 26 Chromatogram (m/z 191) of sample 30994 from Dosariyah.

The most important molecular ratios to source bitumen samples, are Ts/Tm and Gammacerane/C_{30}. These molecular ratios were plotted together with those of other archaeological samples in order for further data interpretation (Figure 27). First of all, it is noticeable that most of the bitumen samples cluster together, indicating that the bitumen from Dosariyah shares the same origin. There are a couple of outliers though. Due to a very low-quality chromatogram Ts and Tm of sample 28266 could not be determined, a problem attributed to biodegradation. The Gammacerane/C_{30} ratio however is in line with those from the bulk of the samples, indicating a similar origin (although it is impossible to say anything conclusive on this sample without its Ts/Tm value). Samples 26531 and 26551 have a noticeable higher Gammacerane/C_{30} ratio, but this might be attributed to differential alteration of the hopane compounds due to biodegradation. The Ts/Tm and Gammacerane/31R ratios on the other hand indicates that these samples probably belong to the main cluster of samples. This main group of samples correlates best with the Ubaid 3-period bitumen from Oueili, but they also are to be located in the molecular vicinity of samples from several Ubaid 3-period samples from Kosak Shamali (bitumen from this dataset also came from the Hit area and the Samsat seepage in Turkey) and Ra’s al-Jinz. All of the samples located within the main cluster of the dataset have been attributed to one or several seepages located in northern Iraq (Connan et al., 1996, Connan and Nishiaki, 2003, Connan et al., 2005). In the case of Dosariyah, the al-Fattha seepage is the most likely candidate as supplier for this bitumen.

These results are in accordance with the observations made based on the $\delta^{13}C$ values, and the somewhat deviant sample 27083 is in this analysis securely located in the main cluster of the measured values, indicating that this sample also has a northern Iraqi origin. Figure 28 is a graphical representation of both techniques, and confirms the close relation between the Dosariyah samples and the northern Iraqi-origin samples from Oueili, Kosak Shamali and Ra’s al-Jinz. This hypothesis was also confirmed by using a statistical approach to the data; the values used for this can be found in Table 3.

Samples 29197 and 29239 are notable two outliers in the dataset (see Figure 27)which appear to have a higher correlation with the H3/as-Sabiyah samples. The chromatogram of sample 29239 had a higher level of background noise than any other sample in the dataset, probably caused by biodegradation. And although this may have influenced molecular ratios to alter, we do not consider it likely that this alteration could have been significant in this sample. The measured $\delta^{13}C$ (-27.75‰) contradicts the information derived from molecular ratio’s and is reminiscent of the seepages in northern Iraq.

Sample 29197 on the other hand did not have a noteworthy background noise enabling reliable peak identification and –calculation. Also the statistical approach clusters this sample together with those from H3/as-Sabiyah, indicating it might originate from the Burgan Hill seepage. It is however conspicuous that exactly these two samples are notable outliers, as they were both recovered from the oldest deposits at the archaeological site, and the group of bitumen from which they belong are well separated from bitumen samples from other layers (see Figure 22, samples 29197 and 29239 are the two white triangles at the bottom). Based upon the molecular data from

GC-MS analyses and the fact that both samples have been excavated from the same archaeological context, we would be inclined to treat them the same and allocate them to the same seepage. It is difficult to say anything conclusive on these two samples, we therefore opt to treat them as outliers in the dataset.

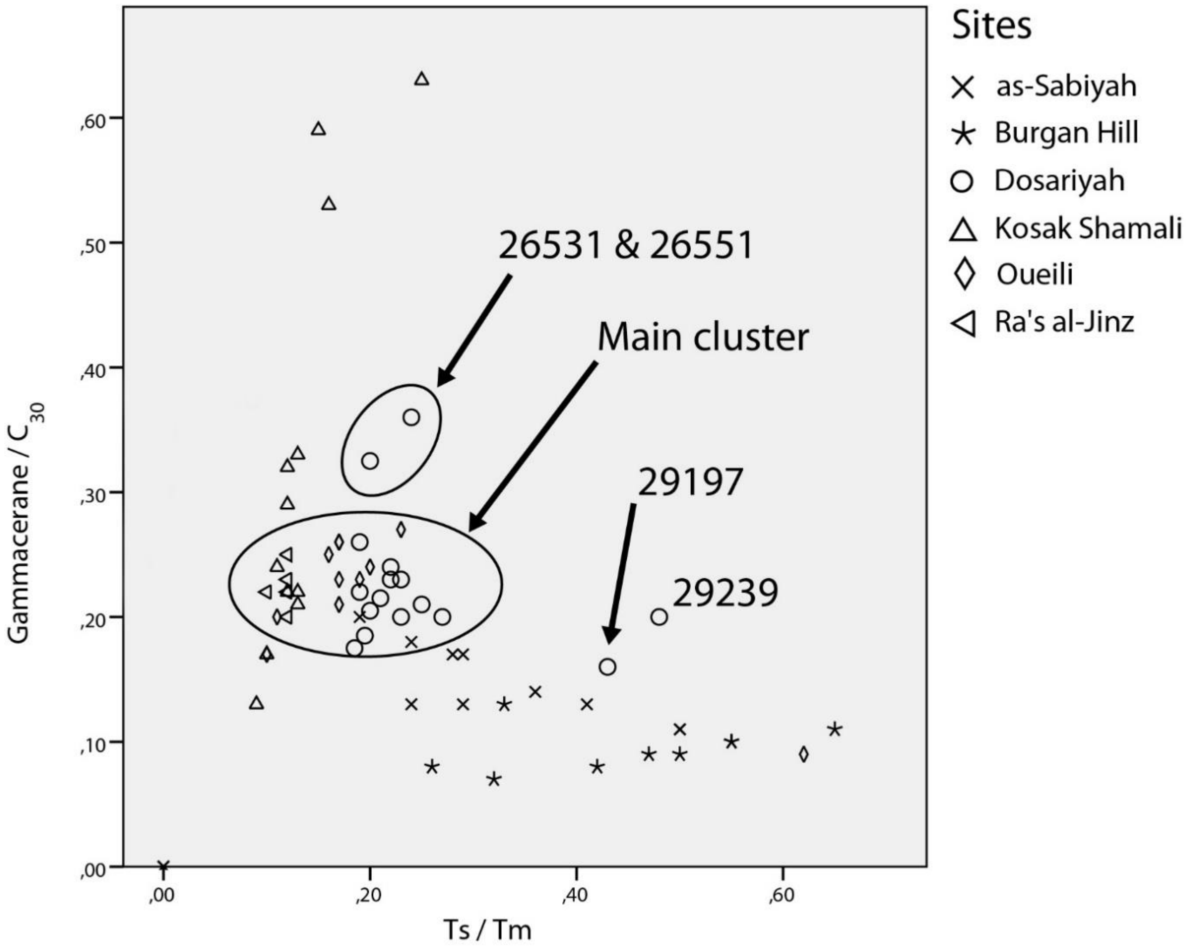

Figure 27 Plot of Ts/Tm vs. Gammacerane/C_{30} hopanes. This graph shows the molecular ratios of bitumen from several archaeological sites and seepages. The main cluster represents bitumen from northern Iraq from various sites; whilst clear outliers are the bitumen from H3/as-Sabiyah & Kosak Shamali.

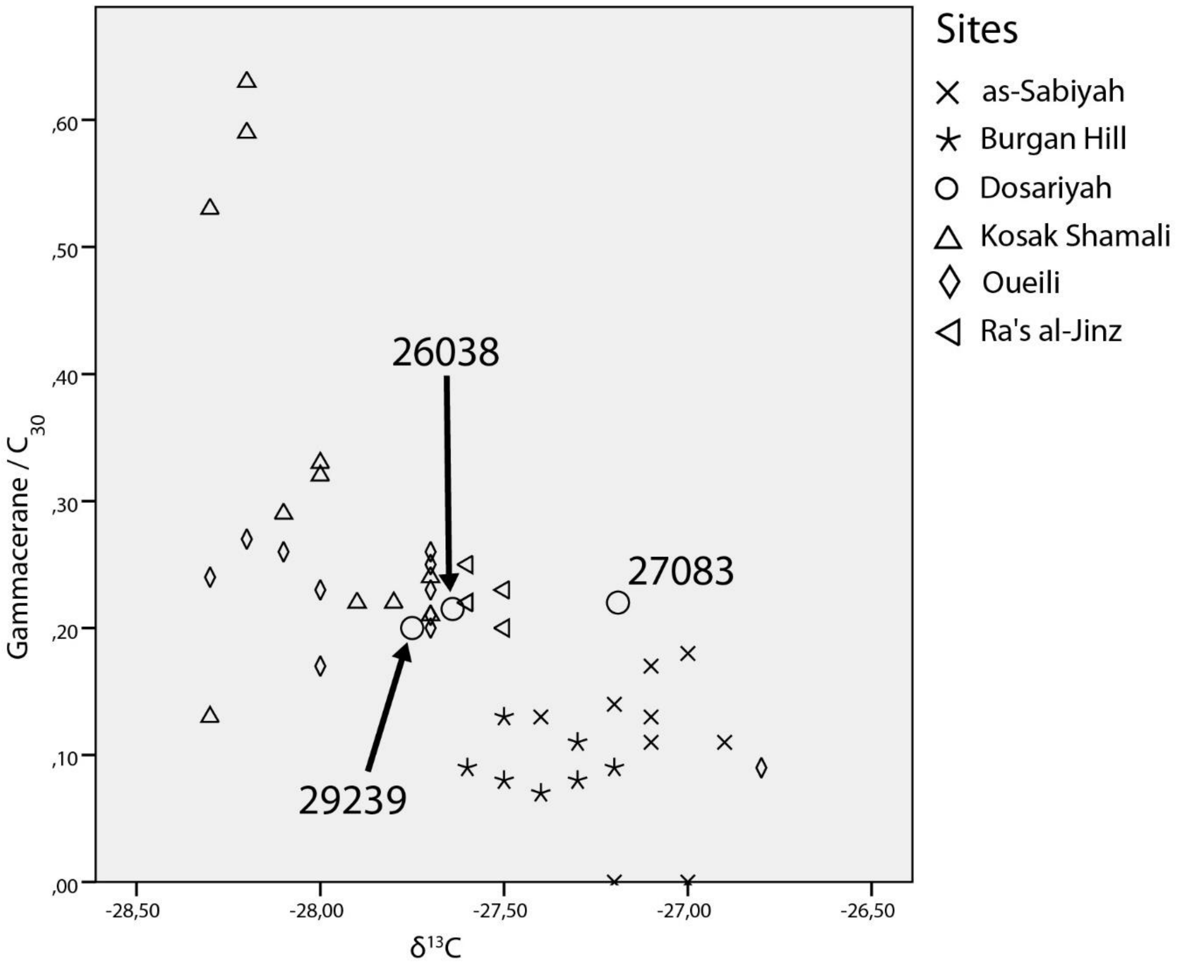

Figure 28 Plot of $\delta^{13}C$ vs. Gammacerane/C_{30}

Table 3 All measured data from the Dosariyah and reference sites used in this research. The data from the reference sites is reproduced from Connan 2010, Connan et. al. 1996, Connan et. al. 2005, Connan & Nishiaki 2003. The sample ID's from Dosariyah correlate with the find ID's, whilst those of the other sites are identical to the ones used in the above-mentioned literature.

Site	Sample	Dating	$\delta^{13}C$	Ts/Tm	GCRN/C_{30}	GCRN/31R
Dosariyah	14330	5000-4500		0.20	0.19	0.52
	14668	5000-4500		0.27	0.20	0.55
	16190	5000-4500		0.23	0.20	0.57
	19554	5000-4500		0.22	0.23	0.61
	25364	5000-4500		0.23	0.23	0.54
	26038	5000-4500	-27.64	0.21	0.22	0.65
	26341	5000-4500		0.20	0.21	0.64
	26531	5000-4500		0.20	0.33	0.52
	26551	5000-4500		0.24	0.36	0.60
	26607	5000-4500		0.19	0.26	0.67
	27083	5000-4500	-27.19	0.19	0.22	0.57
	28266	5000-4500		/	0.22	0.49

	28814	5000-4500		0.25	0.21	0.54
	29197	5000-4500		0.43	0.16	0.50
	29239	5000-4500	-27.75	0.48	0.20	0.45
	30852	5000-4500		0.19	0.18	0.60
	30994	5000-4500		0.22	0.24	0.68
as-Sabiyah	1644	5300-4900	-27,4	0,24	0,13	
	1645	5300-4900	-27,1	0,5	0,11	
	1646	5300-4900	-26,9	0,5	0,11	
	1647	5300-4900		0,19	0,2	
	1648a	5300-4900	-27	0,24	0,18	
	1649a	5300-4900	-27,1	0,29	0,17	
	1649b	5300-4900		0,28	0,17	
	1649c	5300-4900		0,29	0,13	
	1650	5300-4900	-27			
	1771	5300-4900	-27,2			
	1772	5300-4900	-27,1	0,41	0,13	
	1773	5300-4900	-27,2	0,36	0,14	
Oueili	125	5600-5000	-26,8	0,62	0,09	
	21	5000-4500	-27,7	0,17	0,26	
	22	5000-4500	-27,7	0,17	0,21	
	23	5000-4500	-27,7	0,11	0,2	
	28	5000-4500	-28,2	0,23	0,27	
	29	5000-4500	-28,3	0,2	0,24	
	32	5000-4500	-28,1	0,17	0,26	
	34	5000-4500	-28	0,1	0,17	
	37	5000-4500	-27,7	0,16	0,25	
	40	5000-4500	-27,7	0,17	0,23	
	42	5000-4500	-28	0,19	0,23	
Ra's al-Jinz	757	2500-2300	-27,6	0,1	0,22	
	759	2500-2300	-27,6	0,12	0,25	
	762	2500-2300	-27,6	0,12	0,22	
	770	2300-2100	-27,5	0,12	0,2	
	771	2300-2100	-27,5	0,12	0,23	
Kosak Shamali	1375	4940-4550	-27,7	0,11	0,24	
	1376	5100-4700		0,1	0,17	
	1377	5100-4700	-28,3	0,09	0,13	
	1378	5100-4700	-28,3	0,16	0,53	
	1379	5100-4700	-28,2	0,15	0,59	
	1380	5190-4800	-28,1	0,12	0,29	
	1381	5190-4800	-28	0,13	0,33	
	1383	5300-4900	-27,7	0,13	0,21	
	1384	5300-4900	-27,8	0,12	0,22	
	1385	4550-4260	-28,2	0,25	0,63	
	1386	4550-4260	-28	0,12	0,32	
	1395	5300-4900	-27,9	0,13	0,22	

4.1.4 Discussion

Bitumen was a natural resource quite commonly used in the Near East in antiquity, and has been found on many sites especially in Mesopotamia and Iran, where bitumen seepages were plentiful. It has been established that most bitumen used on archaeological sites —especially those located in Mesopotamia, Iran, and the Persian Gulf— came from either one of the three major zones of extraction: the Hit area in central Iraq, the Mosul area in northern Iraq, and the seepages in southwest Iran around the Deh Luran and Susiana Plain (Connan, 2012, Connan and Van de Velde, 2010). Although current bitumen seepages are known from Bahrain and Oman, this bitumen has never been attested in archaeological samples and all bitumen from archaeological contexts derives from at least one of the three above-mentioned zones (Connan and Carter, 2007, Connan and Mouton, 1999, Connan, 2011, Connan et al., 1998, Connan, unpubl.). The only seepage in the Gulf that was in use during Antiquity appears to have been the Burgan Hill (Kuwait). Bitumen from that seepage has been used at the Neolithic site of H3/as-Sabiyah (Kuwait) and the Bronze Age site F6 at Failaka island (Connan and Carter, 2007, Connan, 2010, Connan et al., 2005)

The site of H3/as-Sabiyah was the first Neolithic site in the Gulf from which bitumen was analysed, and now Dosariyah complements this. It is remarkable that the seepage from which the bitumen from H3/as-Sabiyah and Dosariyah conspicuously differs, whereas both sites can be placed in comparable archaeological contexts. Nevertheless, both the radiocarbon dating and the spectrum of painted pottery from H3/as-Sabiyah suggest an occupation of this site predating the settlement at Dosariyah.

This chronological difference might explain the difference in the origin of the bitumen. At the South-Mesopotamian settlement of Oueili, a sequence of bitumen covering a span of stratigraphic layers was analysed. And although we realize the potential pitfall of generalizing an entire area based upon one site, we here accept Oueili as exemplary for south Mesopotamia, mainly due to lack of any other bitumen analyses for this area and period. During the Ubaid 0 to -2 periods at Oueili bitumen from southwest Iran was imported to and used at the site, whereas this seems to change in favour for northern Mesopotamian bitumen at the Ubaid 3 period (Connan et al., 1996). The Ubaid 3-period at Oueili is dated contemporary with the main occupation at Dosariyah and it should therefore be no surprise that the type of bitumen used at both sites shares the same origin.

During the Ubaid 3 period a phenomenon can be observed that has been described as the 'Ubaid expansion' (Oates, 1993, Oates and Oates, 2004, Stein and Özbal, 2007). A decorative style of monochromic "*black on buff*" pottery, but also other aspects of material culture, architecture and ideological structure reminiscent to Southern Mesopotamian Ubaid, can be found within an area stretching from the Mediterranean to the Caucasus and the Gulf. This wide distribution suggests the emergence of a vast interaction sphere driven by exchange networks and cultural interchange (Stein, 2010). The appearance of bitumen from northern sources in southern Mesopotamia and beyond likewise coincides precisely with the development of the “northern Ubaid” (Stein, 1994: 44). The presence of boat models at inland sites in northeast Syria, central Iraq, and eastern Iraq also suggest the emergence of a regular and intensive riverine exchange at that time (Carter, 2012: 352). It is therefore plausible to argue that southern Mesopotamia gained increasing access to northern bitumen sources during the Ubaid 3 period. If the black-on-buff pottery found at Dosariyah was imported from southern Mesopotamia, bitumen could have easily travelled to the Central Gulf in the same way.

But why then the difference between H3/as-Sabiyah and Dosariyah, if both sites share a common cultural context? As stated above, H3/as-Sabiyah predates Dosariyah, and was probably inhabited whilst settlements in Mesopotamia still imported their bitumen from Iran. Quite possibly, bitumen was regarded as a precious commodity due to a certain scarcity or irregular supply from local sources, making it not profitable to redistribute it further. That would force the inhabitants of H3/as-Sabiyah to look for other sources of bitumen, which they apparently found at Burgan. As bitumen at H3/as-Sabiyah has been used for the caulking of boats (Carter, 2010: 91-99), considerable demand of this material can be expected. But locally available bitumen might also have been scarce as well, as the bitumen found at H3/as-Sabiyah all clustered in and around the same building as if they were stored there for later re-use (Carter, 2010: 100-101) indicating value and/or scarcity.

This hypothesis could also help explain the geochemical signature of samples 29197 and 29239 in the dataset from Dosariyah: Both samples come from the lowest deposits and can therefore be considered as representing the oldest bitumen samples from the site. That would suggest that Dosariyah didn’t receive its bitumen from Mesopotamia but was forced to either obtain it directly from a seepage, or through trading partners and –networks. Analyses have shown that the most likely source of these 2 bitumen samples is the Burgan Hill, not surprisingly as we have evidence of the extraction of this seepage from H3/as-Sabiyah. Even the possibility that this bitumen was imported through H3/as-Sabiyah is not unlikely, as analysis of the pottery from Dosariyah indicates a chronological overlap.

Another aspect to consider is the difference in applications for which bitumen was used. At H3/as-Sabiyah, bitumen was mainly —if not exclusively— used to coat boats to waterproof them and make sure they didn’t get waterlogged, a practice quite commonly known from both archaeology and epigraphy (Connan and Carter, 2007, Cleuziou and Tosi, 1994, Carter, 2006, Carter, 2012, Stol, 2012, Vosmer, 2001). This type of specific usage is commonly defined by the presence of barnacles on the bitumen, and as of yet, no such bitumen was found at Dosariyah. The bitumen artefacts show a wide variety of uses; stoppers, plugs, small vessels, possibly as a coating for woven vessels or mats (to be used in architecture). In general, we are comparing a specific usage to a very wide array of applications. The limited usage of bitumen at H3/as-Sabiyah could of course also indicate scarcity, as people used the limited amount of bitumen only for the most necessary of applications, in this case as a supporting role of the maritime network.

4.1.5 Conclusions

This research aimed to provenance bitumen samples that were excavated at Dosariyah. Out of the 20 samples that were used for analyses, three were discarded for further data interpretation due to unreliable data from weathering and biodegradation. Seventeen samples have been investigated using GC-MS analysis on the Saturated Hydrocarbon fraction, of which three were subjected to GC-MS analysis. Results from similar types of research formed the frame of reference to correlate the archaeological samples to bitumen seepages. Fourteen of these samples were successfully related to the bitumen seepages in northern Iraq, with two other samples with questionable origin. One of those two samples is probably related to the Burgan Hill seepage (Kuwait), whilst another one could either be from Kuwait or northern Iraq as both analytical techniques contradict each other in this.

Dosariyah is the second Neolithic site from which bitumen was extensively investigated, and the data and results described in this paper form therefore a major contribution to our understanding of early trading networks in the Persian Gulf. Dosariyah yielded a considerable amount of imported Mesopotamian Ubaid black-on-buff pottery, so it should in the first place not be a surprise that its bitumen is also imported from Mesopotamia. This stands in stark contrast with H3/as-Sabiyah, where locally available bitumen from the Burgan Hill was used, primarily for the caulking of boats. We would suggest the difference in dating as the major factor in explaining the difference in bitumen supply; a remarkable shift in bitumen supplier is to be seen in the archaeological bitumen from Oueili (South Mesopotamia) around the Ubaid 3 period. Possibly, bitumen from northern sources became plentiful in south Mesopotamia only at the advent of the Ubaid 3 period, contemporaneous with the expansion of the Ubaid interaction sphere. As a consequence, the fact that H3/as-Sabiyah slightly predates Dosariyah suggests that bitumen was harder to get and not from northern Mesopotamia but through alternative sources at that time. The possible identification of two bitumen samples from the Burgan Hill in the oldest deposits from Dosariyah does not contradict this hypothesis, but may indicate that both settlements overlap in their occupation, meaning that bitumen was also exploited at the Burgan Hill, or brought in from H3/as-Sabiyah. Only with the emergence of a vast interaction sphere during the Ubaid 3 period, communities in southern Mesopotamia gained access to northern bitumen sources. From this time onwards, northern Iraqi bitumen found its way into the Central Gulf area.

4.1.6 Acknowledgments

The authors would like to thank Dr. Ali al-Ghabban, Assistant Secretary General at the Saudi Commission for Tourism and Antiquities, for the possibility to study the Middle Neolithic site of Dosariyah and its archaeological remains within the framework of the joint Saudi-German Dosariyah Archaeological Research Project (DARP). Work at Dosariyah would not be possible without the generous support by Jamal Omar, Saudi Commission for Tourism and Antiquities in Riyadh. We would like to thank him for his ongoing interest and advice to this project. Our warmest thanks also go to Mr. Abdulhamid al-Hashash, head of the Dammam Archaeological Museum, for his invaluable help and encouragement to establish the project. We would also like to express our gratitude towards Dr. Jacques Connan for his valuable input and comments whilst interpreting the data from the Dosariyah bitumen.

4.2 Digging into the Ubaid-period bitumen from Dosariyah

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Proceedings of the Seminar for Arabian Studies

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4.2.1 Introduction

Bitumen is a material indigenous to the Near East, especially in Iraq and Southwest Iran. Already in the Chalcolithic period it was very frequently used as an adhesive and waterproof coating. Examples from very different areas are plenty; for example at Hassuna during the Samarra-period where pottery was mended using bitumen (Blackham, 1996: 1), or where grain bins were coated with bitumen to protect them (Maisels, 1993: 98). Also outside of Iraq, bitumen was used commonly at sites such as Chogha Mami (Oates, 1969: 146) and Tepe Farukhabad (Wright and Berger, 1981: 43, 53). All of these sites are located in an area where bitumen seepages are active, it is no surprise then that this material was commonly used at these settlements. Figure 4 shows a map of bitumen seepages which are active today, most of which were already actively exploited in Antiquity. In most cases, bitumen came from one of three main areas of extraction; the Hit area alongside the river Euphrates, the Mosul area in Northern Iraq, and southwest Iran (most notably the area around the Deh Luran- and Susiana plains). Apart from these three areas, also several other seepages were used but generally only supplied sites in the near vicinity. Knowing from which seepage bitumen found on archaeological sites is from may give very valuable information on contacts and trade routes in Antiquity.

4.2.2 Dosariyah: the archaeological site and its Neolithic context

The site of Dosariyah was discovered by Burkholder in 1968 and further explored and excavated by Masry in 1972 (Drechsler, 2011, Masry, 1997). From these excavations, Dosariyah unveiled itself as the largest single occupation site from northeast Arabia for the Arabian Neolithic Period. One of the most remarkable features from the site is the massive amount of Ubaid black-on-buff pottery.

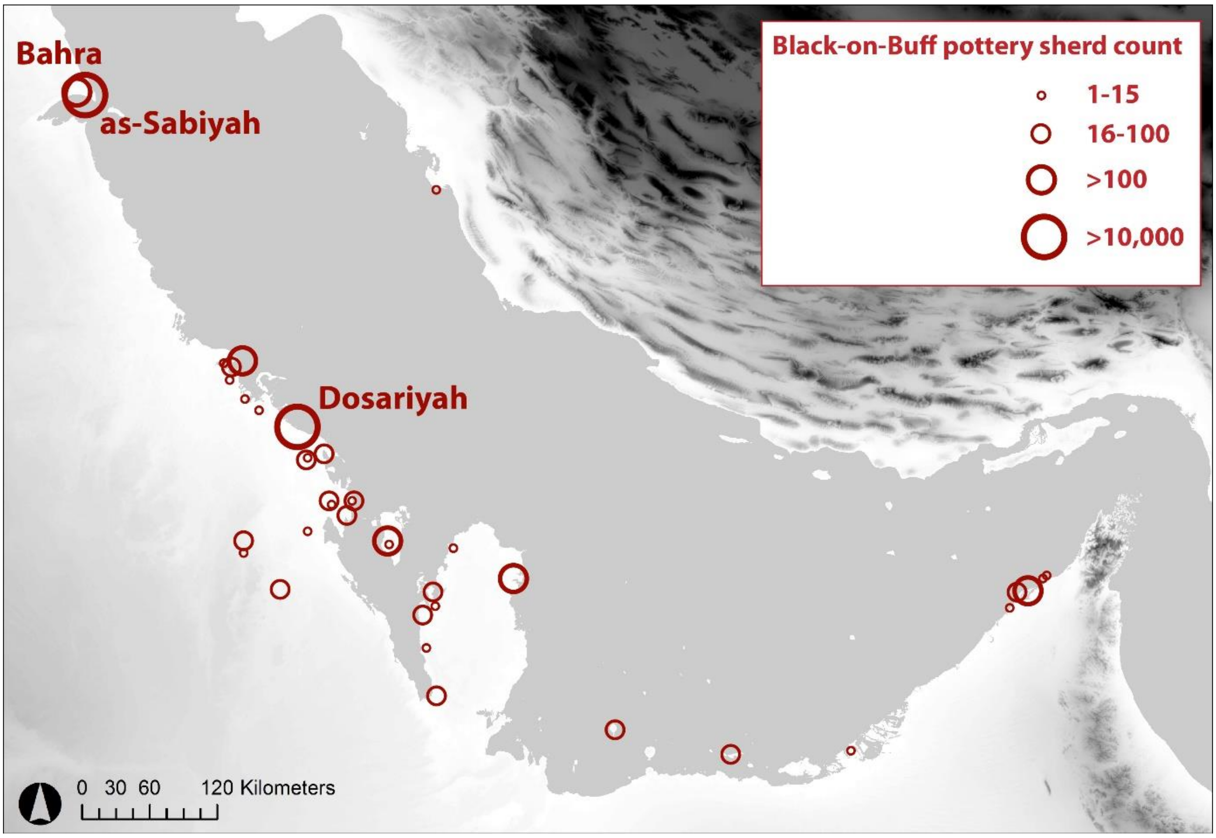

Figure 29 Location of Dosariyah and all the other sites in the Gulf where black-on-buff pottery was found, including an indication on the number of sherds found (modified after Carter & Crawford 2010:3). On this on the following maps, Pournelle's (2003) reconstruction of the Tigris/Euphrates estuary for the relevant period is used.

More recently, the site was under excavation by a joint team from Damman Regional Museum (Saudi Arabia) and the University of Tübingen (Germany). These excavations (and related studies) confirmed the initial dating for the site, placing it somewhere within the first half of the 5th millennium (ca. 5000–4500 BC), and yielded a vast amount of imported black-on-buff pottery and other archaeological materials (Drechsler, 2011:74, 76, 79). Concerning the Ubaid-related pottery, there is only one other site in the Gulf that shows comparable amounts of imported pottery, and that is as-Sabiyah (H3). It is assumed that Dosariyah is one of the major Neolithic sites in the Gulf (see Figure 29).

4.2.3 Analyses of the Dosariyah bitumen

No less than 244 bitumen objects were recovered from Dosariyah, of which many are shapeless lumps or pieces with reed imprints (as bitumen is found more often than not on archaeological sites), but some of them were also real artefacts; such as small bowls and several stoppers (see below). Bitumen was found in almost all of the trenches, although trench E1 held a notably higher amount of this material than any other. Twenty bitumen samples, selected from three trenches and several different stratigraphic layers were sent to Ghent University to determine their geological origin.

Two analytical techniques were used to analyse the bitumen samples. The first of which is Stable Carbon Isotope Analysis, which was conducted on the asphaltenes fraction of the bitumen. The second technique is GC-MS (Gas Chromatography – Mass Spectrometry) and was used in order to identify biomarkers; which are molecular fossils, complex organic compounds consisting of carbon, hydrogen, and other elements. These biomarkers are important as they may reveal a lot of information concerning the geological origin (i.e. the seepage) of archaeological bitumen (Peters et al., 2005a: 3). Four samples were discarded from the dataset due to problems with biodegradation leading to unreliable data. Fourteen of these samples originate from the Mosul are in Northern Iraq, whereas two samples could possibly be related to the Burgan Hill. These two deviant samples could unfortunately not be sourced with the same level of certainty as all other samples. In any case, the main bulk of the samples (14 out of 16) show a clear Northern-Iraqi origin, with a possible association to the Al Fattha seepage. The detailed analytical procedures, including all of the parameters used for interpretation, will appear in Journal of Archaeological Science (Van de Velde et al., 2015) and will also be published in the Dosariyah excavation report (forthcoming).

4.2.4 A bitumen framework

4.2.4.1 Other Arabian Neolithic-period bitumen in the Gulf

In general, very little bitumen from archaeological samples has been analysed, let alone from late 6th – early 5th millennium contexts (see Table 1) . This has to do with the relative low number of sites excavated, the infrequency with which bitumen is found in this area during the Neolithic, but sometimes also with the misidentification of bitumen on-site. This all leads to the fact that bitumen from only two sites has been analysed so far: as-Sabiyah (H3) and Dosariyah (see Table 4).

Table 4 Sites from which bitumen has been analysed for the relevant periods, including the number of samples which were successfully related to their seepage.

Site	Period	Provenance	Sample size
as-Sabiyah (H3)	5300–4900 BC	Burgan Hill	12
Dosariyah	5000–4500 BC	Northern Iraq	14
	5000–4500 BC	Unclear/Burgan Hill?	2
Tell es-Sawwan	5500–5000 BC	Northern Iraq	5
Kosak Shamali	5300–4900 BC	Northern Iraq	3
	5190–4800 BC	Hit	2
	5100–4700	Hit	2
	5100–4700	Samsat	2
	4940–4550	Northern Iraq	1
	4550–4260	Samsat	1
	4550–4260	Hit	1
Oueili	Ubaid 0	Southwest Iran	7
	Ubaid 1	Southwest Iran	14
	Ubaid 2	Southwest Iran	1
	Ubaid 3	Northern Iraq	10

As mentioned before, as-Sabiyah shows some strong parallels with Dosariyah. Fifty-one pieces of bitumen were found at H3, most of which (n=42) showing parallel reeds impressions from the surface of reed bundles, of which 18 had barnacle encrustations on one side (Carter, 2010: 91-98). This evidence indicates that this bitumen was used for the calking of boats, a practice quite common from Antiquity to the modern era. This is not surprising considering the growing importance of boats and maritime networks during this period. Twelve samples from H3 were subjected to analysis, using the same methods as described above, and their results showed that this bitumen came from the Burgan Hill, a small seepage located in Kuwait (Connan et al., 2005, Connan, 2010).

We should however be heedful when comparing the bitumen from H3 and Dosariyah, as it seems to have been applied in different ways at both sites. As most of the bitumen from H3 has been related to seafaring, none of those from Dosariyah seem to indicate a similar usage on first sight. First of all, no bitumen with barnacles attached has been found. Secondly, bitumen was found across the site. This is in contrast to H3, where the spatial distribution of the bitumen indicates that the material had been stored at a specific location, most likely for later re-use (Carter, 2010: 100-101). This is not atypical for settlements in the Gulf. In a similar way, bitumen was also stored at Ra's al-Jinz (Cleuziou and Tosi, 1994: 748), a large collection of bitumen was found inside the warehouse at Umm an-Nar (Frifelt, 1995: 226) and at Ain as-Sayh bitumen was only attested in area C (McClure and Al-Shaikh, 1993). Although none of these sites are contemporary in date, the fact that they all store bitumen in one specific place for re-use, hints at the fact that bitumen was an important and scarce product and preserved for a special purpose, which was more than likely seafaring.

But if the Dosariyah bitumen artefacts were not used for seafaring, what was their usage? Many of the bitumen lumps are reed-impressed, indicating they were the coating of reed mats used in architecture, a quite common usage of bitumen. Evidence of matting used in architecture (for example for roofing) can be found in al-Ubaid (Forbes, 1964: 71), Umm an-Nar Island (Frifelt, 1995: 226), Saar (Moon, 2005: 198) and Southwest Iran (Hole, 1977: 225). Besides that, the bitumen collection also holds several stoppers and plugs, and some small pieces that resemble cups. These cups have very close resemblances to those excavated at the Early Dilmun settlement at Saar. These small beakers consist of a woven vessel, serving as a frame for the bitumen which covered both in- and outside (Moon, 2005: 196). It is very well possible that many shapeless and unrecognizable bitumen lumps were in fact pieces belonging to similar cups, or even larger baskets, which are also very common in the Gulf especially during the Dilmun period on Bahrain and Failaka when they are found on a very frequent basis in the tombs and on the settlements. The same is to say about the corks and stoppers, they appear very frequently on more recent Gulf settlements such Saar, Umm an-Nar, al-Khidr, and Qala'at al-Bahrain (Moon, 2005: 193, Frifelt, 1995: 226, Barta et al., 2008: 125, Højlund and Andersen, 1994: 408). These objects seem to be absent in earlier periods, with the exception of one (probable) stopper from H3 (Carter, 2010: 97). Several types of these objects have been used, but the typologies are not discriminative in place nor time. These objects do not seem to have changed throughout time, indicating they have been around for a long time and serving a very common usage. Similar stoppers are also very common artefacts on Mesopotamian sites (cf. infra).

Although the bitumen objects of Dosariyah have not yet been studied in detail, it is safe to assume that most of them were used in architecture, for the coating of vessels such as cups & baskets, and for the fabrication of stoppers/corks. In general, for a wide array of applications. This stands in contrast with the usage of bitumen at as-Sabiyah, which seem to have been primarily limited to naval purposes. In this case the comparison is between bitumen used for a very specific usage (caulking of boats) to those used for a variety of uses. Could the different in origin of the bitumen of both settlements be related to the different usage of the bitumen? Or could it be more simply a matter of availability? The distance between as-Sabiyah and the Burgan Hill seepage measures roughly 60 kilometers, which is a lot closer than the Mesopotamian settlements in the Tigris/Euphrates Estuary. Nevertheless, the fact that the H3 bitumen was found altogether showing some sort of storage, indicates that the material was not plentiful beforehand. The fact that bitumen was never found at Bahra, a site located in the immediate vicinity of H3, could indicate a scarcity in this area and/or period. However in regard to the fact that bitumen was often stored in specific and unique locations in settlements (cf. H3/as-Sabiyah), it is also very well possible that the excavators at Bahra just didn't touch upon the contexts/locations where the bitumen was stored.

A lot of questions remain unanswered when looking at the bitumen from as-Sabiyah and Dosariyah, and we may ask ourselves that if we were to analyse the bitumen stopper from H3, would that one also have a northern Mesopotamian origin? And consequently, if bitumen boat fragments from Dosariyah would turn up, where would they have come from? In any case, for a better understanding of the bitumen in the Gulf it is interesting to take a look at the bitumen found on Mesopotamian sites.

4.2.4.2 Ubaid period bitumen in Mesopotamia

The work on bitumen from Mesopotamia is slightly more comprehensive for the 6th- to 5th millennium, but in general the dataset remains meagre (see Table 1). We should however take into consideration that many of the southern Mesopotamian sites weren't as systematically and meticulously excavated and recorded as today. That combined with the fact that bitumen is quite prone to disintegration, means that we must grossly underestimate the quantities in which bitumen used to be present at settlements, not just for Mesopotamia but also for the Gulf.

Ubaid 0 to 2

Before turning towards bitumen in the Ubaid 3 period, it might be useful to go a bit further back in time to the Ubaid 0 to 2 periods (roughly 5500–5000 BC). Bitumen from 3 different Mesopotamian sites has been analysed: Kosak Shamali, Tell es-Sawwan, and Tell el' Oueili (see Table 4). The settlement site of Tell Kosak Shamali is located on the Upper Euphrates river and a set of 12 bitumen pieces, covering a timespan from around 5300 BC to 4260 BC has been subjected to analyses. For later half of the 6th millennium the site seems to have had one or possible two (subjected to dating difficulties, see table 1) different supply sources; it is certain that bitumen from Northern Iraq reaches the site, and possibly also bitumen from the Hit source (Connan and Nishiaki, 2003). In the case of Tell es-Sawwan, all five samples (dated 5500–5000 BC.) originate from the Mosul area as well (Connan and Deschesne, 1992b). In the case of Oueili, 41 samples have been analysed of which 22 can be attributed to the Early Ubaid phase (phase 0 up to- and including Ubaid 2). It is remarkable that all of this bitumen originate in southwest Iran (Connan et al., 1996). See Figure 30 for an overview of these sites.

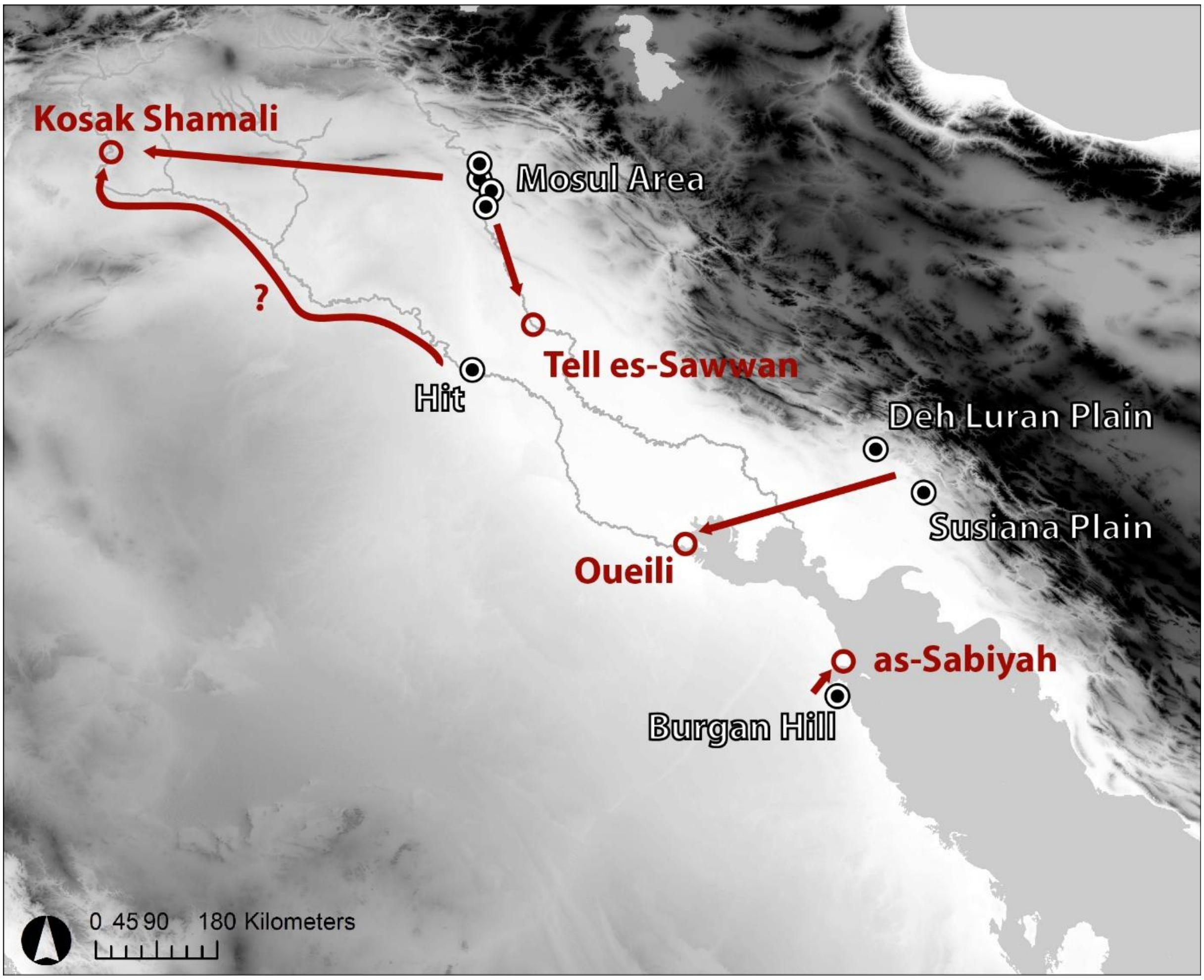

Figure 30 Location of sites mentioned in the text for the Early Ubaid Periods (0 to 2) and the provenance of its bitumen.

When looking at this information, there is one question, which immediately springs to mind: if the inhabitants of as-Sabiyah are 'Ubaidian' in nature (as evidenced by the presence of large amounts of black-on-buff pottery and typical Ubaidian objects, cf. supra), why does this bitumen does not share the same origin than its counterparts from Oueili? The material assemblage of H3 points directly towards Mesopotamian imports, not just for the pottery but also various other smallfinds, would it then be expected that the bitumen would share the same origin as those from Oueili, i.e. Southwest Iran? And if it would be a matter of availability of bitumen from a closer seepage (the Burgan Hill), wouldn't it then be more logical if H3 yielded much more bitumen?

Ubaid 3

For the Ubaid 3 period, during which Dosariyah was inhabited, bitumen from only 2 Mesopotamian sites has been analysed. Again, at Kosak Shamali we see a diversified import of bitumen, as the raw material seems to come from three different sources: northern Iraq, the Hit area, and the Samsat seepage (located in southern Turkey)

(Connan and Nishiaki, 2003). Again, we have to consider that only a handful of samples has been analysed, making it a very small sample size (see Table 4). The other site from which bitumen is analysed, is once more Oueili. Ten samples can be dated to the Ubaid 3 period, and it was concluded that during this period it is bitumen from Northern Iraq reaching the site (Connan et al., 1996), similar to Dosariyah (see Figure 31).

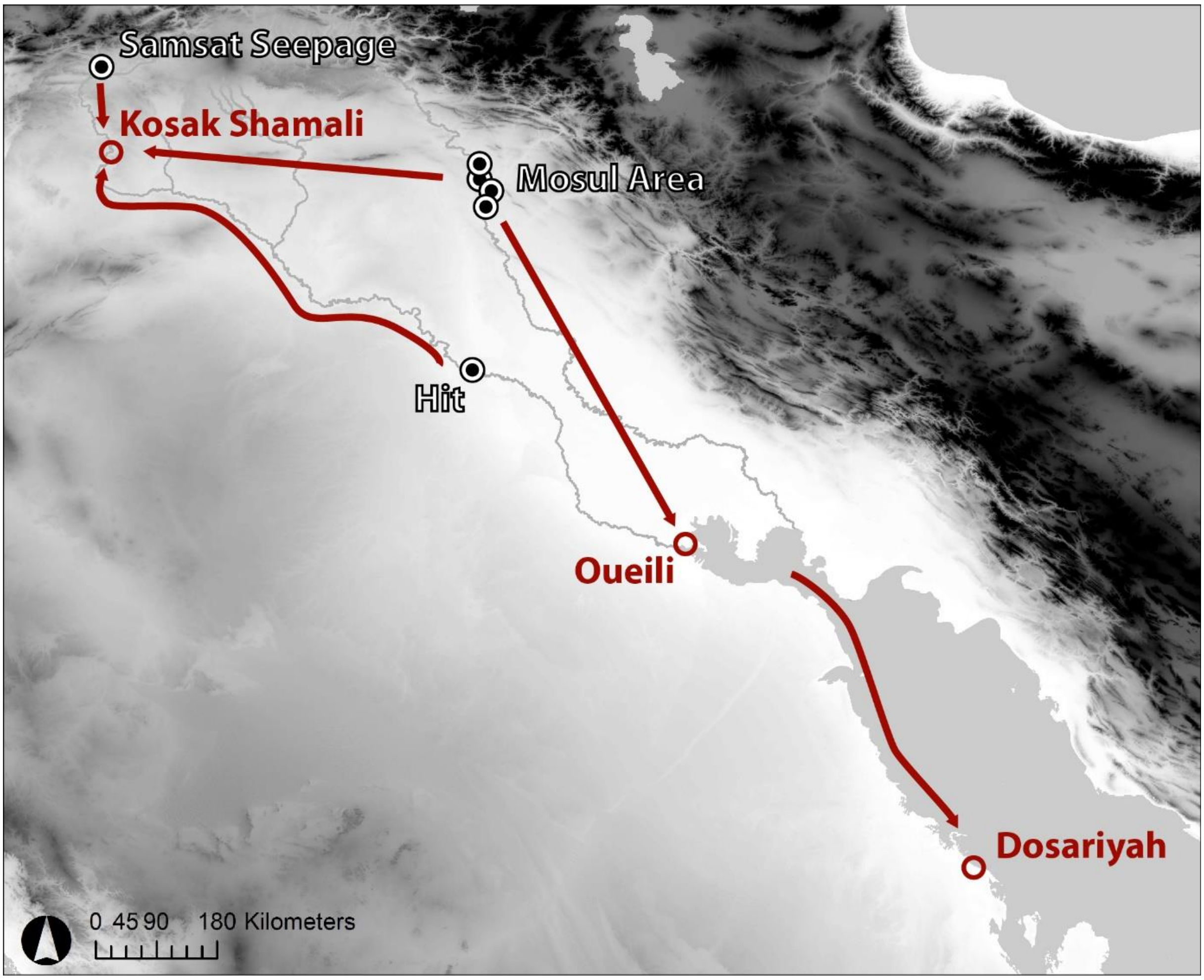

Figure 31 Location of the sites mentioned for the Ubaid 3 period and the provenance of its bitumen.

But what kind of bitumen artefacts are to be found at these Mesopotamian sites? Unfortunately not many bitumen from South-Mesopotamian Ubaid-period settlements has been reported. In the case of Tell el' Oueili there is mentioning of bitumen for different applications and usages; such as matting, small sphere-shaped objects, and other uses (Connan et al., 1996: 414). Unfortunately, no detailed information on the bitumen is available. There is quite some evidence for bitumen at the Ubaid levels at Ur such as basketry, to glue flint in handles, the hair of the famous snake-eyes Ubaidian figurines is bitumen as well in most cases, several lumps, a nail-shaped object, a bitumen ring,... (Woolley, 1955: 9,11,12,92,54,13,68). Also the Ubaid-levels of Uruk yielded several bitumen artefacts, though not as many as the more extensively excavated Uruk levels. We get bitumen used to glue flint in a sickle, two artefacts described as pestles, bitumen forming a handle on flint, and several small bitumen spheres (Lindemeyer and Martin, 1993: 242-244). Not mentioned for the Ubaid period, but matting and stoppers (n=20) have been unearthed from the Uruk levels as well. Several of these stoppers show close resemblance to those of Dosariyah and consequently to other, more recent, examples from settlements in the Gulf (cf. supra).

So similar to Dosariyah, we see that bitumen is being used for the manufacturing of a wide array of common and highly utilitarian products. As contemporary bitumen from many south Mesopotamian sites show a very similar usage, it is no surprise that bitumen from both sites also show the same geological origin.

The Ubaid 3 period is also highlighted by the sudden spread of the typical Ubaid black-on-buff pottery on a large scale, a phenomenon referred to as the "Ubaid phenomenon" (Oates, 1993, Carter and Philip, 2010). The area in which this type of pottery is found ranges from the Mediterranean to the Gulf and testifies of a vast network of both material- and cultural exchange (Stein, 1994: 44). With this expansion, the Ubaid sphere is vastly extended, including Northern Mesopotamia and its bitumen-rich areas in the Mosul area. Most of the economic transactions in the Ubaid 3 period were orientated alongside a north-south orientated axis alongside the rivers Tigris and Euphrates, whereas this axis was rather east-west orientated in the period prior to the Ubaid 3 (Akkermans and Schwartz, 2003: 154). This of course could explain the sudden change in bitumen supplier in Tell 'el Oueili in this period, and consequently also the finding of the same type of bitumen in Dosariyah. Consequently the expansion of the Ubaid to the south is intertwined with that to the north.

4.2.5 Systems of (bitumen) exchange in The Gulf

We established that the bitumen found in Dosariyah have their origin in northern Mesopotamia, and it is logical that after extraction this material was transported over river southwards to settlements in south Mesopotamia such as Uruk, Tello, Ur, Hajji Mohammed and Tell 'el Oueili. Using Pournelle's reconstruction of the Tigris and Euphrates estuary (Pournelle, 2003: 123), all of these sites are located in the mouth of the Gulf-estuary and would have had immediate access to the sea. It is thus most likely that the bitumen found at Dosariyah got there through one or several sites in southern Mesopotamia.

The question that remains concerns the mechanisms of trade and how the bitumen from north-Mesopotamia ended up in Dosariyah. Could it have been through Mesopotamians, who sailed alongside the littoral of the Arabian Peninsula to trade their products at Neolithic settlements such as Dosariyah? A similar way of the dispersal of the Ubaid black-on-buff pottery has been suggested by Oates et. al. (Oates et al., 1977: 232). A down-the-line trade for the spread of Mesopotamian imports, via settlements such as H3 and Bahra seem the most plausible case, as suggested by Carter & Crawford (2010: 208-209). The major issue here however, is that beside Bahra no other ‘Ubaidian’ sites have been identified in the Upper Gulf. Considering the fact that the distance between as-Sabiyah and Dosariyah measures roughly 350 km, as the crow flies, it seems unlikely that seafarers crossed this distance in one trip, without the need of a place of anchorage or re-supply. But unfortunately no other archaeological sites are reported from this area, which is most likely attributed to a hiatus in our knowledge and the archaeological research rather than an actual lack of these settlements. If found, these sites would give very valuable information regarding the systems and mechanisms throughout which Mesopotamian materials found their way all across the littoral of the Arabian Peninsula.

4.2.6 Conclusions

Dosariyah represents only one of the few Neolithic sites in the Gulf where bitumen was found, and is only the second from which bitumen was geochemically analysed in order to identify the origin of this material. The large quantities of bitumen found is remarkable not only because of its quantity —especially in regard to those excavated from as-Sabiyah/H3— but also because of its origin, which is northern Iraq for the majority of the samples.

Remarkably the bitumen from as-Sabiyah/H3, a site which shares many cultural parallels with Dosariyah, came from the Burgan Hill, a bitumen seepage in Kuwait. This difference may be attributed to the fact that analyses was conducted on different types of bitumen; in the case of H3 bitumen was primarily used for seafaring, and in Dosariyah it was used for a variety of uses (such as matting, baskets, stoppers).Another more likely possibility may lie in the fact that H3 is not fully contemporary with Dosariyah. Possibly, bitumen was a rather scarce good in Mesopotamia in the Early Ubaid periods (Ubaid 0 up to -2) making it not beneficial to export it to different regions. This could explain why H3 imports so many goods from southern Mesopotamia, but not its bitumen. The main bitumen supplier for southern Mesopotamia then changes at the Ubaid 3 period, which could have also meant a larger-scale exploitation at the source and easier accessibility to the natural resource, making it more interesting to export. If this was the case, it explains why we don’t find the Northern Iraqi bitumen in the Gulf prior to the Ubaid 3 period. It is however hard to prove this hypothesis, as the nature of bitumen dictates that a quantitative assessment of the material on an archaeological site. This hypothesis, however, offers an explanation as to why bitumen from H3 and Dosariyah differ in origin.

When hypothesizing the entire bitumen networks and the role of Mesopotamia in it, we must take into account though that bitumen from merely a single Southern

Mesopotamian site has been analysed, and it cannot be excluded that bitumen from other seepages also reached south Mesopotamia. Another problem we still face concerns the methods and mechanisms by which bitumen ended up in Dosariyah. We assume the product reaches the Central Gulf through an exchange network via sites in the Upper Gulf. The problem with that, however, is that currently only two sites in that area have been excavated; Bahra and H3, of which on the former no bitumen artefacts were found. However the absence of bitumen from the excavations at Bahra doesn't necessarily mean that it is total absent from the site. As explained above, it is not uncommon for bitumen to cluster on one location on an archaeological site, usually at a warehouse or specific room of a building where the bitumen was stored. Quite possibly the excavators of Bahra didn't touch upon the area or layers where the bitumen is to be found on the ancient settlement.

The bitumen from Dosariyah is only the second Neolithic-period bitumen dataset which was geochemically screened in order to obtain the geological origin of the material. Evidently, these results contribute greatly to our understanding of bitumen trade, but also on a larger scale of Neolithic trading mechanisms and spheres of interaction. There are however still hiatus in our understanding of bitumen trade due to the lack of available datasets.

4.2.7 Acknowledgments

First of all I would like to thank dr. P. Drechsler for entrusting me with a collection of bitumen excavated at Dosariyah, and for supplying me with all kinds of photos and information concerning Dosariyah and its bitumen. I am greatly indebted to prof. F. Lynen, Mike De Vrieze, and Pieter Surmont of the Department of Separation Sciences at Ghent University for support and help with all aspects of sample preparation and GC-MS analysis. I am equally indebted to prof. P. Boeckx, Samuel Bodé and Katia Van Nieuland from Isofys (Isotope Bioscience Laboratory – Ghent University) for their help and support with the stable carbon isotope analysis. I also would like to thank Agnieszka Szymczak for giving me information on Bahra. Finally, I would like to express my gratitude to E. Olijdam and E. Tetlow for their reviews and comments on this text.

Chapter 5 Bitumen in Dilmun

5.1 Sourcing the bitumen from Tell F6 (Failaka)

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Tell F6 on Failaka Island. Kuwaiti-Danish Excavations 2008-2012

Van de Velde, T. Sourcing the bitumen from Tell F6. *In:* *Hø*jlund F. & Abu-Laban A. (eds.) *Tell F6 on Failaka Island. Kuwaiti-Danish Excavations 2008-2012.* Moesgaard: Jutland Arcaheological Society Publications

5.1.1 Introduction

Several bitumen samples from the 2008-2012 excavations in Tell F6 were sent to Ghent for geochemical analysis in order to determine their origin. In the Near East, bitumen surfaces naturally at several locations and raw material from these seepages was extracted by man for a variety of purposes. The material was especially practical as an adhesive but was also used frequently for its waterproofing properties. It has been used in architecture as a mortar or to waterproof reed mats, for the lining of pottery, the assembling of broken pottery, as a material used to make stoppers/corks, to waterproof baskets, to enhance and waterproof the hull of boats etc.

Bitumen samples from several archaeological sites in the Persian Gulf have been sourced in the past, of which most came from the same seepages as the bitumen used in Mesopotamia and Iran (Van de Velde and Bodé, Accepted for Publication, Connan and Carter, 2007, Connan et al., 2005, Connan et al., 1998, Connan and Van de Velde, 2010, Connan, unpubl., Van de Velde et al., 2015). These are to be located in northern Iraq (around Mosul), at Hit (Iraq, middle Euphrates river), the Dead Sea, the Burgan Hill in Kuwait, and a variety of seepages in western Iran (Figure 33).

5.1.2 Bitumen on Bronze-Age Failaka

We can mark the 21st century B.C. as an important era for both Bahrain and Failaka, for it is then that the settlements on the islands start to become major players in the trade- and interregional contacts that characterize the Bronze Age in the Persian Gulf. The fate of the island of Failaka is closely intertwined with that of Bahrain as it is also here, on this strategically-located island in the upper Gulf, that we find strong manifestations of the Dilmun culture. It is especially the work of the Danish pioneering excavations (1958-1963), the following French missions (1983-1988) and the work of the Kuwaiti-Slovak missions (2004-2008) that uncovered the Bronze Age on this island (Calvet and Gachet, 1990, Salles, 1984, Calvet and Salles, 1986, Kjaerum and Højlund, 2013, Højlund, 1987, Benedikova and Barta, 2009, Barta et al., 2008). These teams and excavations worked respectively at the Bronze Age sites of Tell F3, Tell F6, Site G3, and Al-Khidr (Figure 32).

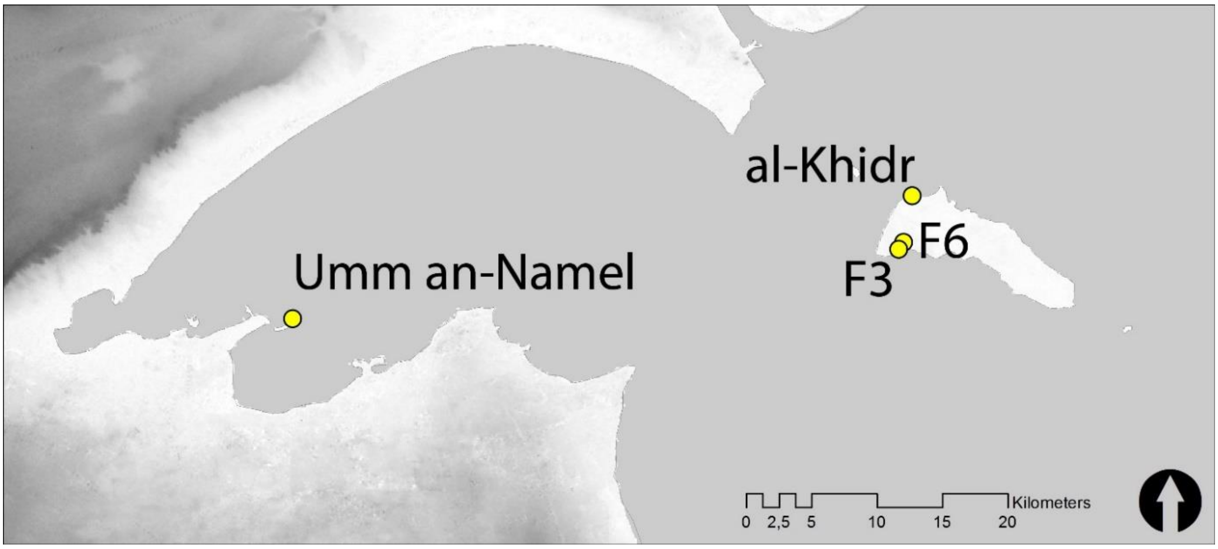

Figure 32 Dilmun-period sites with evidence of bitumen usage.

At all these sites an abundance of bituminous material was found, but the bitumen from the first three of them, Tell F3, Tell F6, and Site G3, has never been studied and is only sparsely mentioned in publications as the following review will show.

In House 26 in Tell F3 (dated c. 1850-1800 B.C.) the plastered surface of a small trough is said to have traces of bitumen (Kjaerum and Højlund, 2013: 81). From Site G3, a dwelling site north of Tell F6, two pottery sherds with traces of bitumen inside are mentioned (Salles, 1984: fig.28:68 & fig.30:92).

More evidence of bitumen presence and –usage is mentioned from the area of the temple in Tell F6 excavated by the French mission and here dated to the end of the 3rd and beginning of the 2nd millennium B.C. Several zones both inside and outside the structure yielded layers with shapeless lumps of bitumen. Unfortunately there is no information available as to what sort of artefacts these bitumen lumps originally belonged, the easily-decaying nature of bituminous material obviously attributes to

that. Beside the bitumen lumps, a "*zone de déchets de bitumen*" is mentioned (locus 389) (Calvet and Gachet, 1990). The excavation report does not give any further information on this context, but it is thus possible that bitumen was worked in the immediate surroundings of the temple.
Bitumen was also attested in close context with pottery (Pic, 2008: catalogue n°'s 8, 43, 140, 168, 229, 241, 249, 250). This could either have been as a lining of the vessels, or be a remnant of the storing/transporting/working of bituminous material.

Noteworthy is a sculptured piece made from a black bituminous stone from the temple at Tell F6 (Calvet and Pic, 1986: 76-77). The material was originally identified as an artificial 'stone' and named "*mastic de bitume*" by O. Deschesne & J. Connan (Connan and Deschesne, 1996). It was, however, later identified as a natural-occurring rock from the Sargelu formation in Iran (Connan, 2012:156-177) and should not be considered as actual bitumen.

It is clear from the French excavation reports from Tell F6 that bitumen was found frequently on the site. No detailed study on this material has been performed, however, making it hard to make any claims on specific usages or to make an assessment on quantities.

Conversely, excavations at Al-Khidr have revealed a Dilmun-period settlement with massive amounts of bitumen used in a domestic context, and the finds from here have been properly analysed and published (Benedikova and Barta, 2009). Al-Khidr was excavated by a joint Kuwaiti-Slovak team for four campaigns. The site is identified as a dwelling site similar to Tell F3 . All bitumen finds from Al-Khidr have been quantified, registered and —where possible— identified. Around 200-230 kilograms of bitumen was unearthed at the site, including bitumen-coated baskets and cordage, stamp seals, bottle stoppers, or just plain shapeless lumps. There are even occurrences of bitumen layers (largely in contexts with pottery or with stones in walls), in-between building stones as bonding materials, and thick bitumen crusts in large earthenware vessels (Belenová-Štolcová, 2010). The evidence could also point towards bitumen processing on the site, with the many bitumen lumps and –layers in-between the soil matrices and the thick crusts in vessels (which could also be evidence of transporting).

In general there is a lot of evidence for the usage- and working of bitumen at several Bronze Age sites on the island of Failaka, though the material has been unevenly studied and published. The excavations at Al-Khidr showed that bitumen was used for a multitude of purposes, a fact that has been supported by the excavations presented here.

5.1.3 The origin of the bitumen from Failaka

As of yet, only bitumen from the French missions at Al-Qusur, Site B6, Tell F5 and Tell F6 has been analysed in order to obtain information on its geological origin, the latter site being the only Bronze Age site. Geochemical tests have been conducted on thirteen bitumen samples from the Bronze Age levels at Tell F6, three from the Middle Dilmun Period (ca. 1400-1300 B.C.) and ten from the Early Dilmun levels (ca. 2000-1700 BC). All of these samples originated from the Hit area in Iraq, with the exception of one sample which came from the Burgan Hill bitumen seepage (Kuwait) (Connan and Carter, 2007) (Figure 2). Hit-bitumen has been identified at Qala'at al-Bahrain from Middle Dilmun levels, and possibly also from Early Dilmun contexts. There is however also proof that Iranian bitumen was used at the Dilmun settlement at Saar and in several tumuli from the same period (Connan et al., 1998, Van de Velde and Bodé, Accepted for Publication). It seems that bitumen from several seepages and source areas was exported to- and used in several Dilmun settlements simultaneously, although it looks apparent that bitumen from the Hit-seepages was not used in this area prior to 2000 B.C.

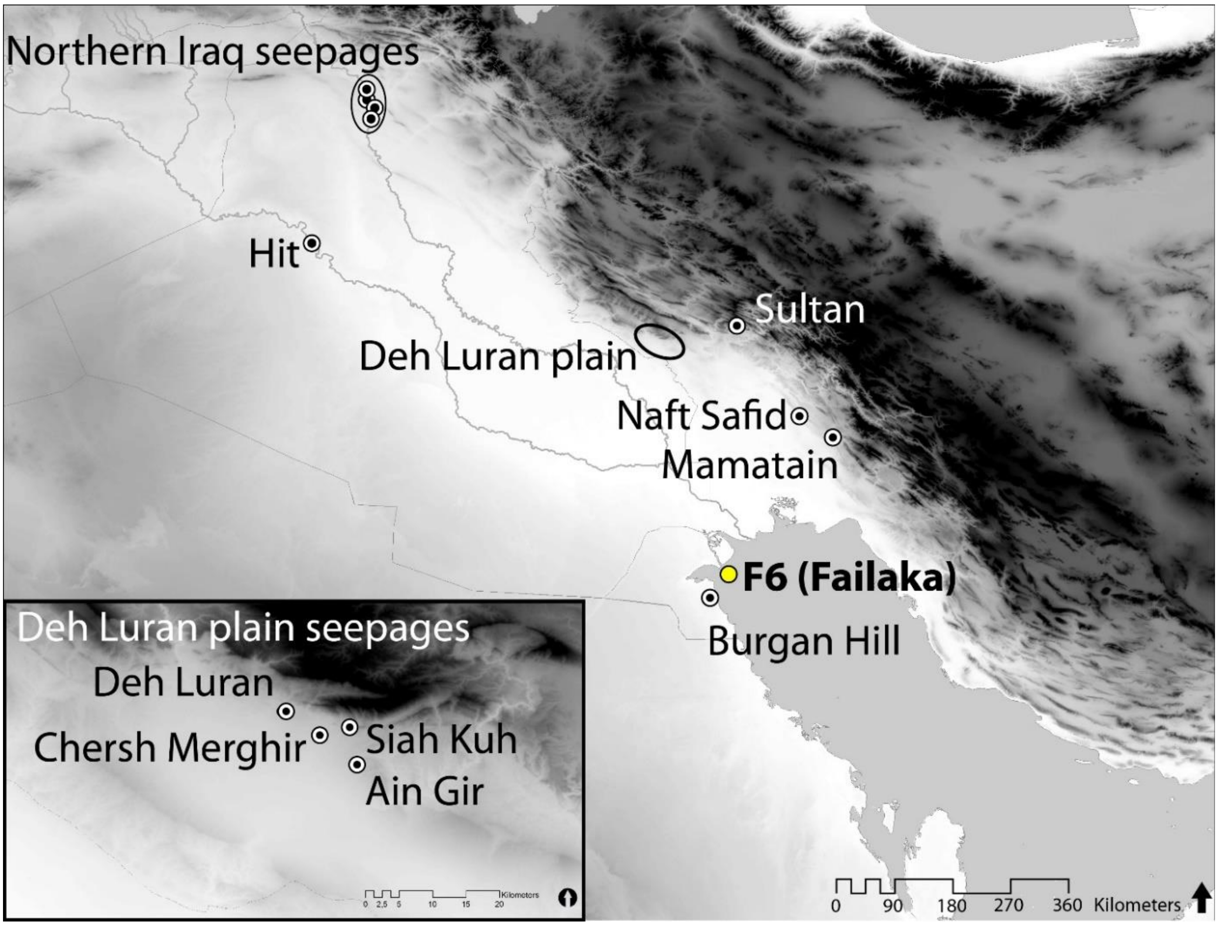

Figure 33 Map showing the bitumen seepages and sites mentioned in this chapter. For a complete overview of all bitumen seepages in Antiquity, see Connan & Van de Velde (2010) and Connan (2012).

5.1.4 Bitumen samples from F6 (Kuwaiti-Danish campaigns)

Thirteen bitumen samples from the 2008-2012 excavations in Tell F6 were selected for geochemical screening to determine their geological origin. Bitumen from several trenches and deposits were chosen in order to investigate whether or not bitumen suppliers changed through time. Faint impressions are visible on several samples, but in all cases these are too small or not enough pronounced to allow any detailed identification.

The viscosity of natural bitumen does not allow the material to be properly worked, therefore a temper was added (Forbes, 1964, Connan and Van de Velde, 2010). These additives were not studied in detail but rather macroscopically checked. There was nothing really noteworthy for most of the samples; most samples had the typical brownish-black colour, often tempered with sand. Samples 9 & 10 however are extraordinary because of the presence of complete shells in their matrix (see Figure 34). Crushed-shell inclusions on the other hand are attested quite frequently in bitumen mixtures, but those should be considered as intrusive material in the initial matrix (i.e. sand) that was used to create a bitumen mixture rather than a deliberate addition. The shells present in samples 9 & 10 have been identified as members of the Potamididae family, a species closely associated with mud flats and mangroves. These shells are only visible on one side of the lumps, in both cases the backside is free of any inclusions, and in one case (sample 9) elongated impressions are visible. These imprints are about 2- to 5 mm wide and probably derive from reed bundles.

Figure 34 Bitumen sample 9, bearing shell inclusions. Note the parallel-impressed lines on the backside (left photo) and the shells on the front.

It is unlikely that shells were added deliberately to the bitumen mixture as that would not enhance its physical characteristics in any way, it would actually make it more porous. So either the shells in this bitumen are an ‘accident’ (for example the spilling of bitumen on a shell-covered surface), or they became part of the bitumen by a physical process. There are strong indications for the usage of bitumen in naval architecture and it is not impossible to imagine shells being impressed into the bitumen coated hulls of boats when they were moored in the mangroves. Bitumen was also a product which was quite prone to re-usage and in some cases there is evidence of the stripping of bitumen from boats with the aim to store these for later re-use (Connan et al., 2005, Carter, 2010). This could also have been the case for the bitumen samples 9 and 10 from Tell F6.

5.1.5 Analytical methods

The same analytical methods as defined in chapter 3.4 were used in this research. Originally, this was all included in the original text to be published in the Tell F6 excavation report, but was deleted here as the methodology has already been outlined before.

5.1.6 The origin of the F6 samples

The geologic origin of the sample was investigated using the information from the terpane fingerprints and the values obtained through stable carbon isotope analysis on the asphaltenes fraction. The $\delta^{13}C$ values of all samples are too high to contain any bitumen from the Hit area. In several samples (samples 1, 3, 4 & 5) 18α-oleanane was observed. The base for this compound is angiosperms and originates in the Triassic Period or earlier, and is for this region mainly linked to oils originating from the Padbeh source-rock formation in Iran (Peters et al., 2005b: 572, Connan, 2012: 117).

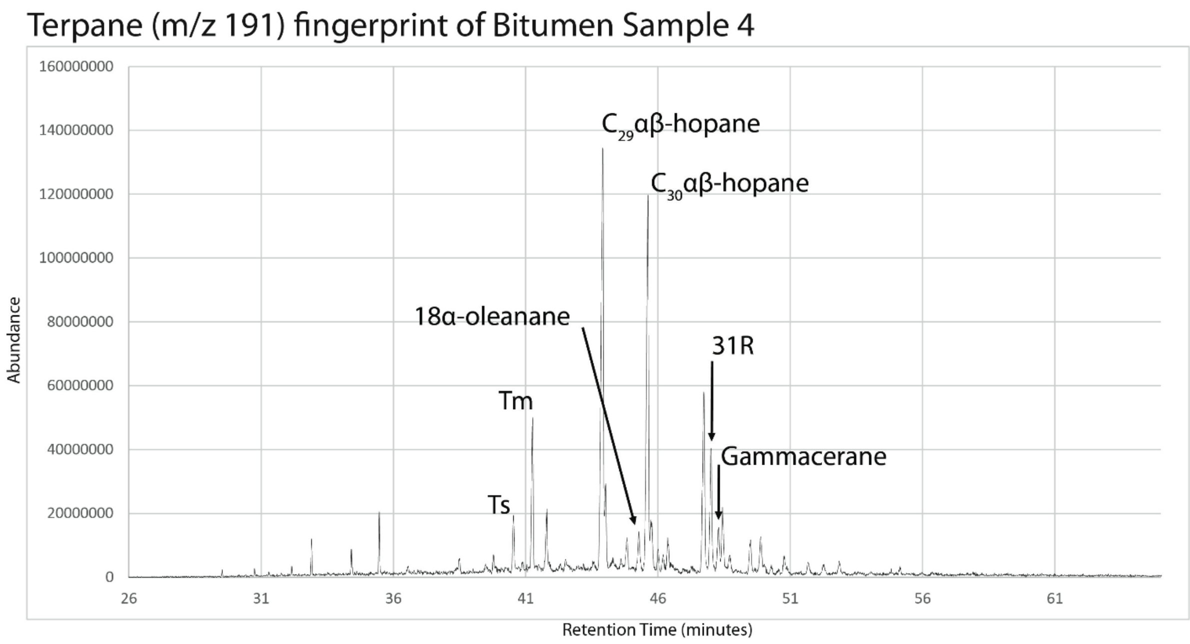

Figure 35 Terpane fingerprint of bitumen sample 4 from Tell F6.

Figure 36 shows several cross-plots combining several parameters; the molecular ratios Ts/Tm and 18α-oleanane/C_{30}αβ-hopane, but also the values retrieved from stable carbon isotope analysis. The oleanane-containing bitumen from Tell F6 seem to cluster nicely together with bitumen samples from the Hellenistic period site of Akkaz, which have been published in detail (Connan, 2011). The exact source of these samples has not been established as their genetic parameters do not match any of the reference seepages, nor any other archaeological bitumen. It has therefore been suggested that this bitumen was either a mixture between two types of bitumen (one from the seepages on the Deh Luran plain mixed with oleanane-containing bitumen) or alternatively this bitumen came from a yet-unidentified seepage (Connan, 2011). We should however mention that the Tell F6 bitumen does not match those from Akkaz on all parameters, the Gammacerane/C_{30}αβ-hopane ratio[5] for instance is remarkably higher in the samples from Akkaz and the range in which the Akkaz bitumen $\delta^{13}C$ values fall is much wider (see Figure 37). Of course, the extreme wide range of $\delta^{13}C$ values in the Akkaz samples is an indication of the presence of bitumen from multiple seepages.

If the bitumen from Akkaz is indeed a mixture of raw material from different seepages (one with a genetic pattern reminiscent of those from the Deh Luran plain and one containing oleanane), it is striking that the 18α-oleanane/C_{30}αβ-hopane ratio is very similar in both datasets. That would indicate that the composition of the mixtures is similar, which would be quite coincidentally considering the 2000 year gap between the two sites. In that respect, it seems more likely that the bitumen from both sites were extracted from an unidentified seepage. The deviating Gammacerane/C_{30}αβ-hopane, the wide $\delta^{13}C$ range of the Akkaz samples, and the fact that as of yet no oleanane-containing bitumen with a $\delta^{13}C$ of asphaltenes within the -28/-27‰ range has been identified in bitumen samples (Connan, 2011) on the other hand conflict with this hypothesis.

From the ten bitumen samples which have been subjected to GC-MS analyses, only 4 showed a presence of oleanane. The same genetic parameters as described above were used to identify the source of the other 6 samples. Both molecular ratios and $\delta^{13}C$ match with bitumen from Susa, which lacks oleanane and has been identified as coming from one of the seepages on the Deh Luran plain[6]. Bitumen from the Burgan Hill shows a very similar molecular pattern (Connan, 2010, Connan et al., 2005), but as illustrated in figure 6, the $\delta^{13}C$ of this bitumen does not match with the values from the Tell F6 samples. Consequently, we can securely state that the Tell F6 bitumen samples without oleanane are coming from one of the seepages on the Deh Luran plain. The same type of bitumen

[5] It should, however, be mentioned that this parameter is considered to be less reliable than the others.

[6] This is true for a part of the Susa dataset, some bitumen samples of this dataset do contain oleanane, and several have also been identified as coming from Sultan (Connan 2012: 127).

has also been identified in archaeological bitumen samples from Bronze Age tumuli in the A'ali burial field in Bahrain (see chapter 5.2) and possibly also in the archaeological bitumen samples from Saar Settlement (Connan et al., 1998).

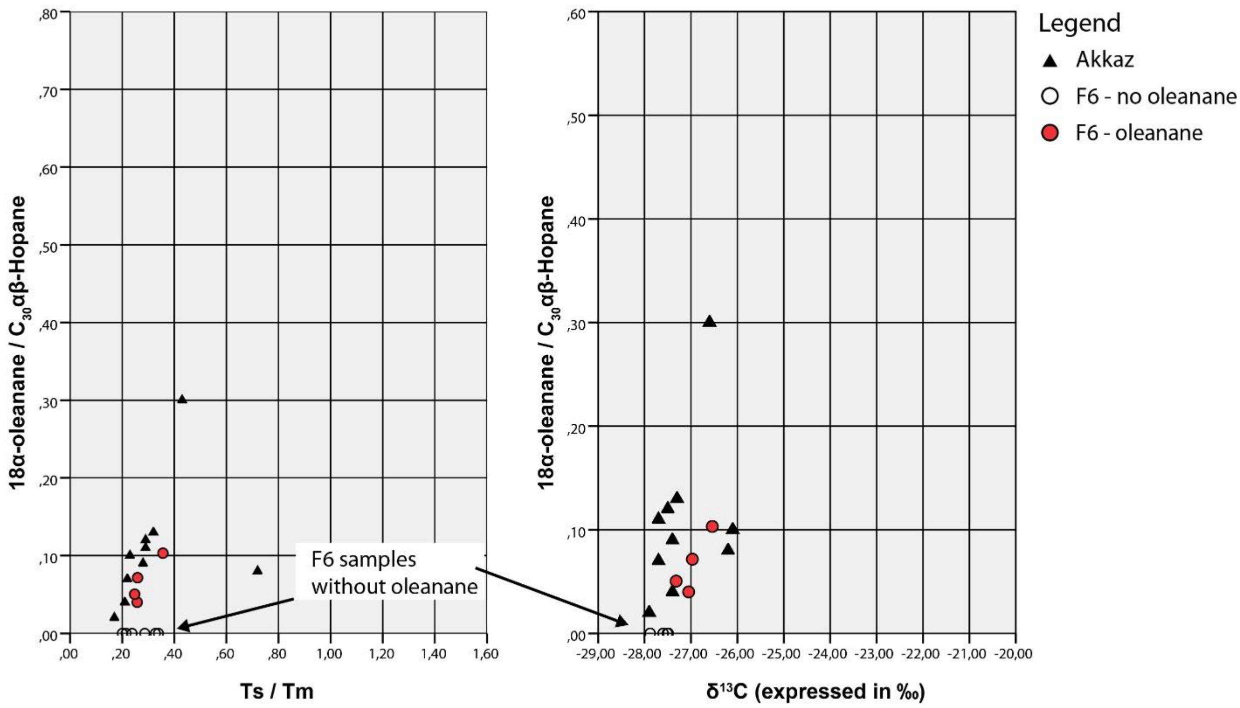

Figure 36 Cross-plots of Ts/Tm vs. 18α-oleanane/C_{20}αβ-hopane (left) and δ^{13}C vs. 18α-oleanane/C_{20}αβ-hopane (right). The bitumen samples from Tell F6 containing oleanane seem to correlate with those from Akkaz (molecular ratios and δ^{13}C retrieved from Connan 2011).

If the bitumen containing oleanane are mixtures, then it would make sense that one of the components is material from the seepages on the Deh Luran plain as the molecular parameters (Ts/Tm, Gammacerane/C_{30}αβ-hopane, Gammacerane/C_{31}22R hopane) between the two groups show great similarities (Figure 37). There is, however, a distinction between the two groups based on the δ^{13}C measurements, which are lower in the samples without oleanane. This, however, does not necessarily mean anything as this value of course could be influenced by the addition of bitumen from another seepage.

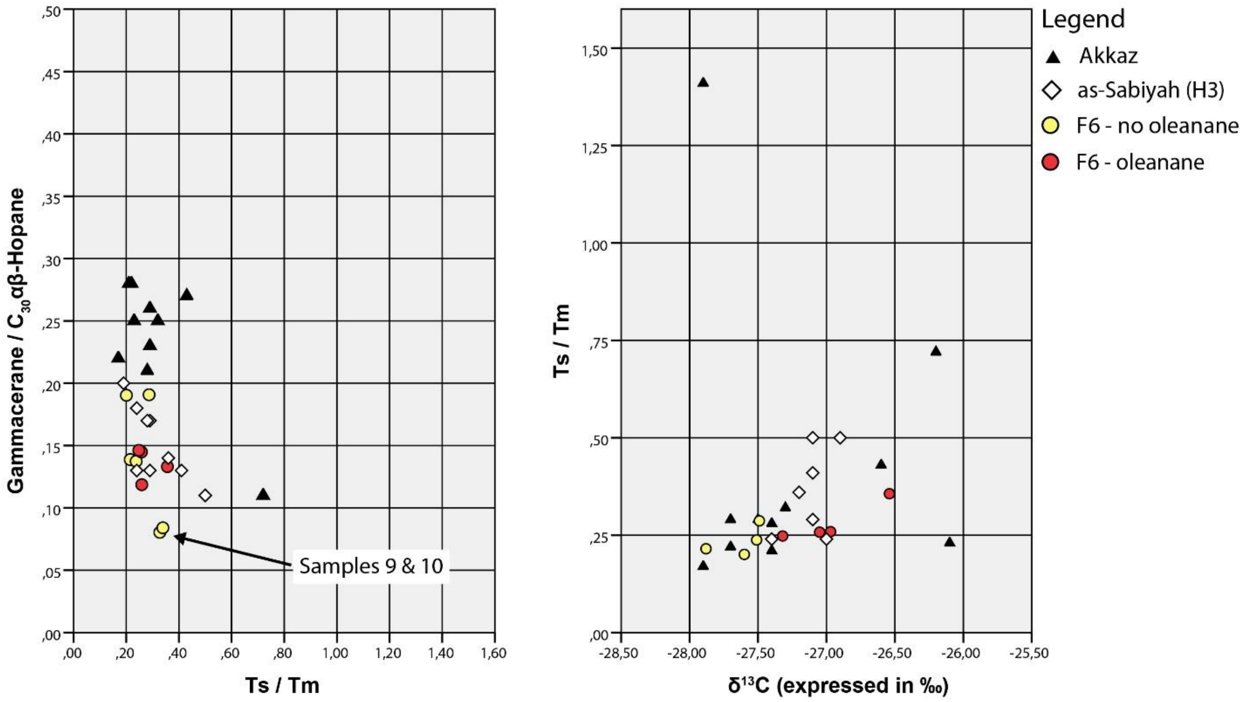

Figure 37 Cross-plots of Ts/Tm vs Gammacerane/$C_{30}\alpha\beta$-hopane (left) and $\delta^{13}C$ vs. Ts/Tm (right). The bitumen samples excavated at the Neolithic site of H3/as-Sabiyah have been identified as coming from the Burgan Hill (Kuwait).

For three samples no molecular data was retrieved and only $\delta^{13}C$ is available. The values of samples 12 & 13 are both low in number (-27,8‰ and -27,83‰) and correlate with the $\delta^{13}C$ of the samples coming from the Deh Luran plain. Sample 11 on the other hand shows a relatively low $\delta^{13}C$ (-26,77‰) in such a degree that it falls within the range of the samples containing oleanane and should therefore probably be placed in the same group.

Samples 9 & 10 are outliers in the dataset (see Figure 37) and do not seem to belong to either of the two groups defined above. These two samples do not exhibit any oleanane and are characterized by their low Gammacerane/$C_{30}\alpha\beta$-hopane. Alteration of the Gammacerane/$C_{30}\alpha\beta$-hopane parameter is possible because of biodegradation of the sample. Unfortunately no isotopic data is available for these samples. Either this bitumen comes from an unidentified seepage, or we are encountering a chemically alteration of the samples. It is remarkable that these samples do not only distinguish themselves from the rest of the dataset because of their molecular ratio, but also because of the high number of shell inclusions (cf. supra). It remains most likely that this bitumen shares the same origin as the other non-oleanane containing samples, i.e. one of the seepages on the Deh Luran plain.

Table 5 Measured values of $\delta^{13}C$ and molecular ratios used in this research.

Sample	$\delta^{13}C$	Ts/Tm	GCRN/C_{30}	GCRN/31R	OLN/C_{30}	Source
1	-27,05	0,26	0,14	0,41	0,04	Mixture?
2	-27,88	0,22	0,14	0,4	/	Deh Luran plain
3	-26,97	0,26	0,12	0,36	0,07	Mixture?
4	-26,54	0,36	0,13	0,4	0,1	Mixture?
5	-27,32	0,25	0,15	0,47	0,05	Mixture?
6	-27,5	0,2	0,19	0,51	/	Deh Luran plain
7	-27,49	0,29	0,19	0,49	/	Deh Luran plain
8	-27,51	0,24	0,14	0,42	/	Deh Luran plain
9		0,33	0,08	0,24	/	Deh Luran plain?
10		0,34	0,08	0,25	/	Deh Luran plain?
11	-26,77					Mixture?
12	-27,8					Deh Luran plain?
13	-27,83					Deh Luran plain?

5.1.7 Bitumen sample J18

One specific feature that was excavated at Tell F6 was a stone set pit with a very fine-grained black deposits at the bottom (Figure 38). It was though that this pit was a fire pit specifically for the heating- and processing of bitumen, hence the black colour of the lower deposit, and a sample was taken for geochemical screening. GC-MS on this sample revealed a very obscure chromatogram lacking all the distinctive peaks identified in bitumen. However, $\delta^{13}C$ on this sample gave a value of -27,32‰, which correlates nicely with that of bitumen. Previous experimental work on bitumen revealed that the recycling —or more specifically, the reheating— of bitumen caused the evaporation of entire compound classes, yet that the isotopic signature on the asphaltenes remains the same independent of the duration of the reheating of the sample (Hollander and Schwartz, 2000). If the fire-pit discussed here was indeed used as an installation for the working of bitumen, its contents would naturally be subject to numerous processes of (re-) heating, causing the archaeological sample to behave like it does on the chemical analyses. So quite possible, the black deposit in the stone set pit is the silent witness of bitumen processing, but we should not take this for granted as no bitumen-specific molecules could be identified. Unfortunately, the experimental work discussed above does not include detailed chromatograms and consequently it was impossible to compare those results with that of sample J18, nor was it possible in the current research to duplicate similar test.

Figure 38 The stone set pit from which bitumen sample J18 was recovered (black deposit at bottom of pit).

5.1.8 Conclusions

Thirteen bitumen samples from the 2008-2012 excavations at the Bronze Age site of Tell F6 on Failaka have been investigated with the aim to determine their original seepage. GC-MS analysis was conducted on ten samples, and stable carbon isotope analysis on eleven. Using these analytical techniques, it was possible to identify two different main groups in the bitumen dataset, discriminated by either the absence or presence of oleanane – a chemical molecule only present in bitumen coming from the Padbeh source rock formation (Iran). The group of samples containing oleanane shows close resemblance to a group of bitumen samples from the Hellenistic site of Akkaz. No specific origin for these samples was given, but it was suggested that this bitumen was either a mixture of raw materials from different sources, or coming from an unidentified seepage. The former option being the most plausible. The other group of bitumen samples from F6, lacking oleanane, is coming from one of the bitumen seepages on the Deh Luran plain (Iran). The dataset knows two true outliers, samples 9 & 10, for which no reference data was found. It is most likely that this bitumen is also coming from the Deh Luran plain.

As Iranian bitumen has been identified in several Dilmun-period datasets from both Failaka and Bahrain, it should come as no surprise that this type of material is also found at Tell F6. Remarkable, however, is the difference in bitumen between the Tell F6 bitumen datasets from the French- and the Kuwaiti-Danish missions. Most of the material from the French excavations of the temple is Iraqi in origin (Hit-area), whereas one sample was identified as coming from the Burgan Hill seepage (Kuwait).

5.1.9 Acknowledgments

I am indebted to Prof. F. Lynen, M. De Vrieze and P. Surmont from the Department of Separation Sciences at Ghent University for their support and help concerning the GC-MS analyses. The same goes for Prof. P Boeckx, S. Bodé and K. Van Nieuland from Isofys (Isotope Bioscience Laboratory – Ghent University) concerning the stable carbon isotope analysis. I would also like to express my gratitude towards Prof. D. Van Damme (Palaeontology Department – Ghent University) for identifying the shells in bitumen samples 9 & 10. Last but not least I would like to thank J. Connan for his support and valuable comments on this research.

5.2 Analysis of bitumen from the Royal Mounds (Bahrain)

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Laursen S.T (ed.) *The Rise of Kingship and the Early Dilmun State in Bahrain.*
Moesgaard: Jutland Arcaheological Society Publications

5.2.1 Introduction

Bitumen was sampled from a number of different archaeological contexts in connection with the recent investigations at the 'Royal Mounds of A'ali (this vol. Chapter 3). In the framework of the current project, these bitumen samples were analysed in order to determine the geological origin of the bitumen.

The samples came from five different burial mounds of which 'Mound A', 'Mound E', 'Mound N' and 'Mackay Tomb 29' are interpreted as Royal tombs proper and 'Mound D' is considers the tomb of a high ranking "aristocrat". The bitumen had in each case been used in connection with the construction of the chambers with the exception of 'Mound E' where it presumably had been used for lining a basket. In 'Mound A' the bitumen was found on palm mats that have been inserted between two corresponding courses on both sides of the chamber wall. The mats are interpreted as "mudflaps" aimed at directing water away from the chamber. 'Mackay Tomb 29' produced bitumen samples from palm mats found in the chamber walls which probably also were the remains of some type of rain diverting "mudflaps". The bitumen from 'Mound D' was sampled from a continuous layer of palm mats which had been placed in the mound fill as a huge "umbrella" over the two super imposed chambers. Here the function of the bitumen again appears to have been to divert rainwater from the chambers. In 'Mound N' the bitumen samples were taken from palm mats which had been place on the contact surface between the capstones of the chamber and the walls which supported the chamber roof. The bitumen coasted mats in 'Mound N' is considered to have acted as a caulk which too was intended to keep the chamber dry.

5.2.2 Early Dilmun period bitumen

Bitumen has been found at various other archaeological sites in the Persian Gulf, especially in Dilmun-related contexts. In Early Dilmun period contexts (c.2300-1750 B.C.) bitumen has been attested frequently in Bahrain not only at the settlements at Qala'at al-Bahrain (Højlund and Andersen, 1994, Højlund and Andersen, 1997) and Saar (Moon, 2005), but also in burial mounds (Højlund, 1995) and the Barbar temples (Andersen and Højlund, 2003). In Kuwait, bitumen was found in large numbers at the sites of Tell F6 (Calvet and Gachet, 1990, Kjærum and Højlund, 2013, Højlund, 2012; 1987), al-Khidr (Benedikova and Barta, 2008, Barta et al., 2008, Belenová-Štolcová, 2010) and Umm an-Namel (Connan and Carter, 2007). This evidence strongly indicates that bitumen was a product commonly used by the inhabitants of the islands of Failaka and Bahrain and that it must have been available in substantial quantities. The wide-spread usage of bitumen in architecture —as found in Bahrain and Failaka— also points to that conclusion (Connan and Van de Velde, 2010) (Figure 39).

All the bitumen found in Dilmun-related contexts was imported from several seepages, as discovered by analysis on archaeological samples from the sites of Qala'at al-Bahrain, Saar, Karranah, Buri, Umm an-Namel, Tell F6[7], and Bahrain Tumulus B-5[8]

[7] Two different datasets have been subject to geochemical analysis, one from the French excavations (Connan & Carter 2007), and one from the Kuwaiti-Danish Archaeological project (Van de Velde, in preparation)

(Connan et al., 1998, Connan and Carter, 2007, Van de Velde, Accepted for Publication-a). These analyses established that multiple seepages were exploited and their raw materials exported to Dilmun. They further document a probable shift in the supplier of bitumen to Dilmun around 2000 B.C. The bitumen found in the early layers at Qala'at al-Bahrain (period 2500-2000 B.C.) and Umm an-Namel (2100-2000 B.C.) seem to have originated from the seepages in northern Iraq, whereas in the later phases at Qala'at al-Bahrain and Tell F6 bitumen from Hit has been identified (Connan and Carter, 2007, Connan et al., 1998). Additionally, bitumen from southwest Iran has also been identified in samples from Tell F6, Saar and several Bahraini tumuli (Connan et al., 1998, Van de Velde, Accepted for Publication-a). It should, however, be noted that although the separation line appears to fall around 2000 B.C., this is based on a low sample size. Only eleven samples from a total of three sites is to be placed prior to 2000 B.C.: one from Saar, five from Qala'at al-Bahrain and five from Umm an-Namel.

Quite remarkably Hit bitumen is the only type found in the later Kassite period (samples dated 1400-1300 B.C.) both at Failaka Tell F6 and Qala'at al-Bahrain (Connan and Carter, 2007), and in this way the appearance of Hit bitumen in the early 2nd millennium contexts must be considered as heralds. The forces driving the change in bitumen distribution after the 21st century B.C. is most likely Mesopotamia rather than Dilmun.

5.2.3 Archaeological samples

In the past, mainly bitumen from settlement contexts has been analysed, whereas only five bitumen samples from three different burial contexts have been investigated; the burial sites being Karranah, Buri and Tumulus B-5 (exact location unknown). The origins of the five samples from Burial contexts are to be found in Iran (in case of Karranah and Buri) and probably Iraq (in the case of tumulus B-5) (Connan et al., 1998, Van de Velde, Accepted for Publication-a). The current dataset expands this list by adding 10 bitumen samples from 5 different tumuli from the A'ali burial field.

The five tumuli from which bitumen has been sampled are Mound A, D, N, E, and Mackay Tomb 29 (Table 6). All samples should be dated somewhere in the 2000-1750 B.C. timespan, with 'Mound A' 'Mound D' and 'Mackay Tomb 29' falling in the early half and 'Mound E' and 'Mound N' falling in the later half. The majority of the bitumen was sampled from palm mats that had been used in the construction of the royal tombs with

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[8] This sample comes from the bitumen lining of a vessel deposited in tumulus B-5, and excavated by Peter B. Cornwall (Van de Velde, in preparation).

the exception of the samples from 'Mound E'; this bitumen was probably part of a bitumen coated palm basket deposited in the tomb.

Table 6 Bitumen samples taken from the A'ali Burial Field (Bahrain).

Sample N°	Mound	Comments:
Sample 1	Mound A	From "mudflaps" in chamber east wall
Sample 2	Mound A	From "mudflaps" in chamber east wall
Sample 3	Mound A	
Sample 4	Mound A	From "mudflaps" in chamber west wall
Sample 5	Mound A	From "mudflaps" in chamber west wall
Sample 6	Mound N	
Sample 7	Mound N	From bitumen coated palm mat coated on south chamber wall cantilever
Sample 8	Mound D	From "umbrella" in fill over chamber
Sample 9	Tomb 29	From possible "mudflaps" in chamber wall
Sample 10	Mound E	From possible bitumen coated basket

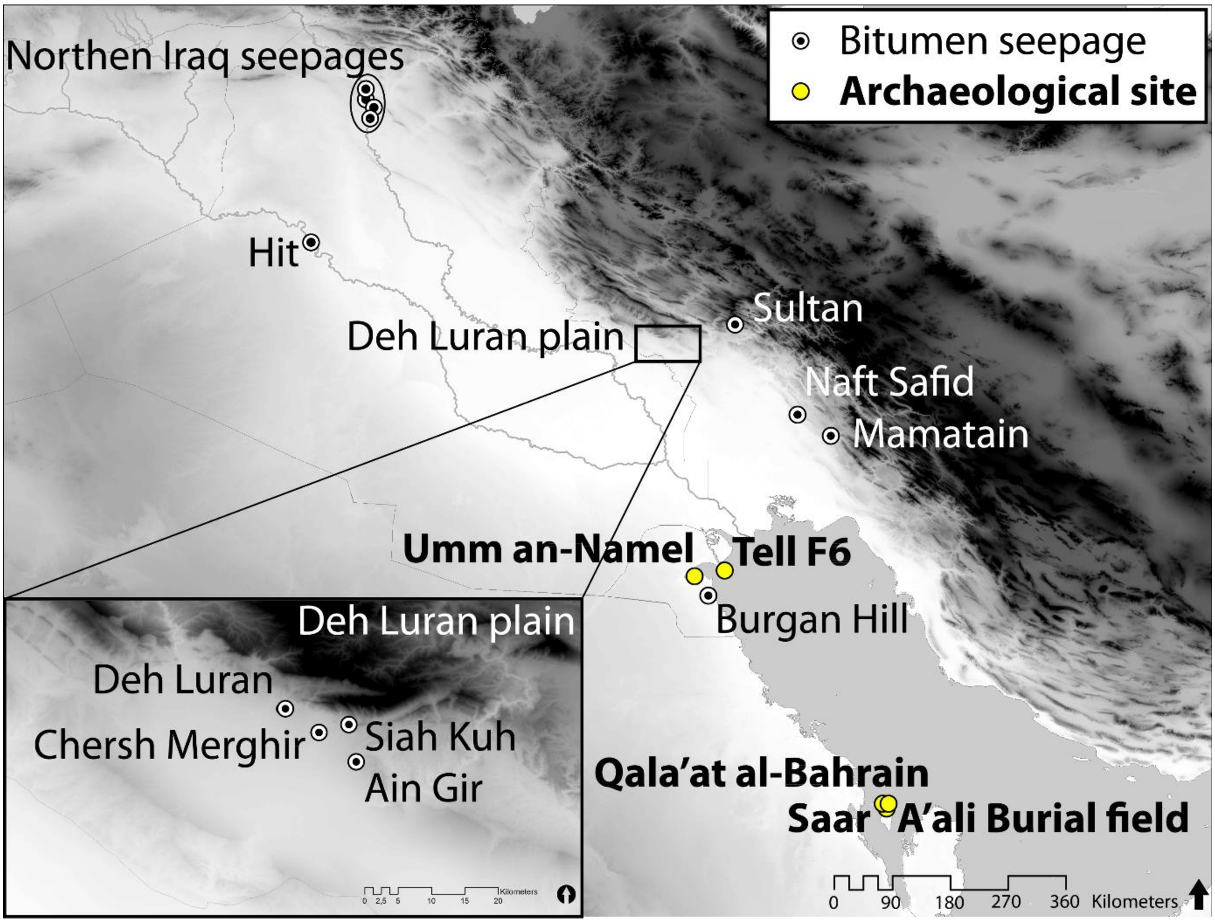

Figure 39 Map showing the most important active bitumen seepages in Antiquity and the major archaeological sites mentioned in this chapter.

5.2.4 Analytical techniques

The same analytical techniques as discussed in chapter 3.4 were used. Although the chemical protocol is included in the original publication of this dataset, it will not be repeated here.

5.2.5 Analysis of the data

5.2.5.1 Primary Dataset characteristics

Figure 40 shows a cross-plot between molecular ratios of Ts/Tm and Gammacerane/$C_{30}\alpha\beta$-hopane. The data shows two apparent small clusters, and two obvious outliers. The data retrieved from Carbon Isotope Analysis doesn't shed any more light on these clusters, as all measured values fall within the same range (-27,7 ± 0.1 ‰, Table 7). We should however bear in mind that not all samples underwent this type of analysis, and it is unfortunate that there are no $\delta^{13}C$ values from the two extreme outliers (samples 5 & 10).

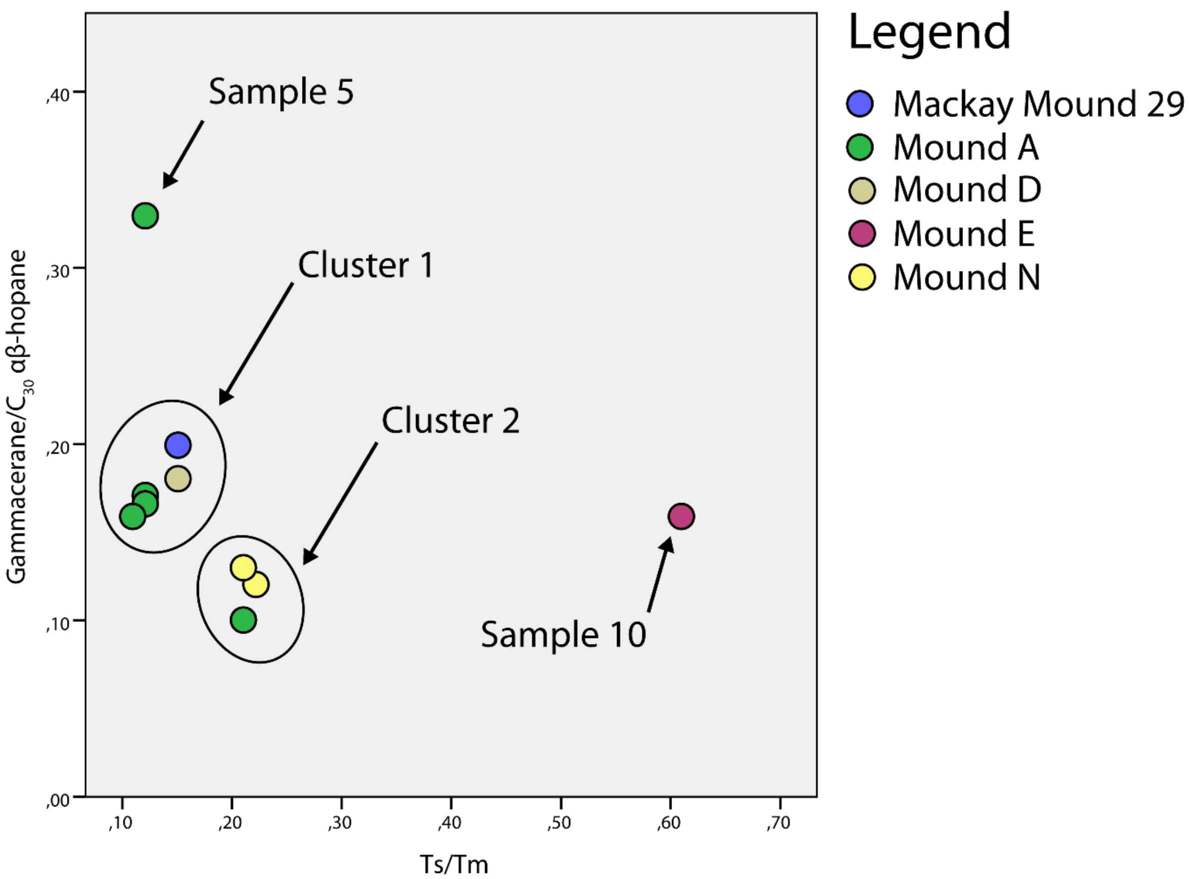

Figure 40 Ts/Tm vs. Gammacerane/$C_{20}\alpha\beta$-hopane cross-plots. The bitumen seem to form two clusters with two outliers, samples 5 & 10.

The majority of the samples from 'Mound A', and those of 'Mound D' and 'Mackay Mound 29' cluster together, whereas both samples from 'Mound N' seem to form another cluster together with one sample from 'Mound A'. If we accept that these clusters represent different seepages, then we have strong indications that bitumen from both areas was used simultaneously. The sample from 'Mound E' (Sample 10) should be considered as having a separate origin than the other samples, rather than being an outlier. The presence of 18α-oleanane, which was not observed in any other sample, attribute to that. The base for this compound are angiosperms and originate in the Triassic Period or earlier, and is for this region mainly linked to oils originating from the Padbeh source-rock formation (Peters et al., 2005b: 572, Connan, 2012: 117). Accordingly, this bitumen sample can be regarded as an import from a seepage located in southwest Iran. Oleanane has also been attested in archaeological samples from Susa, Abu Chizan, Tall-e Geser, and Tell F6 (Failaka) (Connan, 2012, Connan et al., 2008, Connan et al., 2014, Van de Velde, Accepted for Publication-b).

5.2.5.2 Sourcing the A'ali bitumen

Molecular ratios from Abu Chizan and Tall-e Geser were compared with those of the A'ali samples (Figure 41). The 18α-oleanane to C_{30}αβ-hopane ratio of A'ali bitumen sample 10 (value of 0,3) falls within the range of the samples from Abu Chizan, as do the Ts/Tm and Gammacerane/C_{30}αβ-hopane ratios. This bitumen has been identified as a composite oil from the Lower Cretaceous Middle Khazdumi and the Eocene Padbeh source rocks, and coming from either the Mamatain- or Naft Safid oil seeps (Connan et al., 2008). The bitumen sample from 'Mound E' probably also comes from one of these two seepages. For an overview of the Iranian bitumen seepage and their 18α-oleanane to C_{30}αβ-hopane ratio, see Figure 20.

The other outlier in the dataset (Sample 5, from 'Mound A') shows none of the source-specific compounds found in bitumen sample 10. Remarkable from this sample are the low intensities of the C_{30}αβ-hopane peak and the 17α,21β-hopanes (22S and 22R); causing a high Gammacerane to C_{30}αβ-hopane ratio which contribute to this samples outlier position. These oddities are probably related to chemical alteration (biodegradation) rather than a difference in origin of this bitumen.

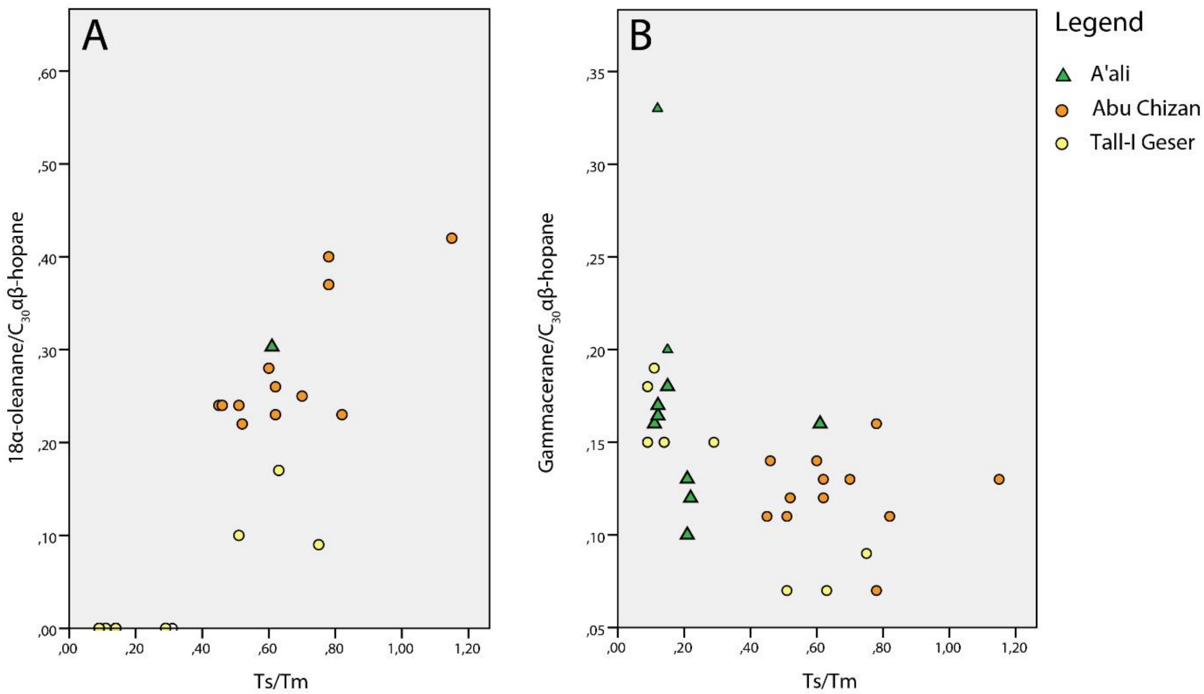

Figure 41 Cross-plots of Ts/Tm vs. 18α-oleanane/C_{30}αβ-Hopane (A) and Ts/Tm vs. Gammacerane/C_{20}αβ-Hopane (B). A'ali bitumen sample 10 (Mound E) matches the samples from Abu Chizan rather than the 3 oleanane-holding samples from Tall-e Geser (other samples from the latter site originate from Sultan and the Deh Luran plain).

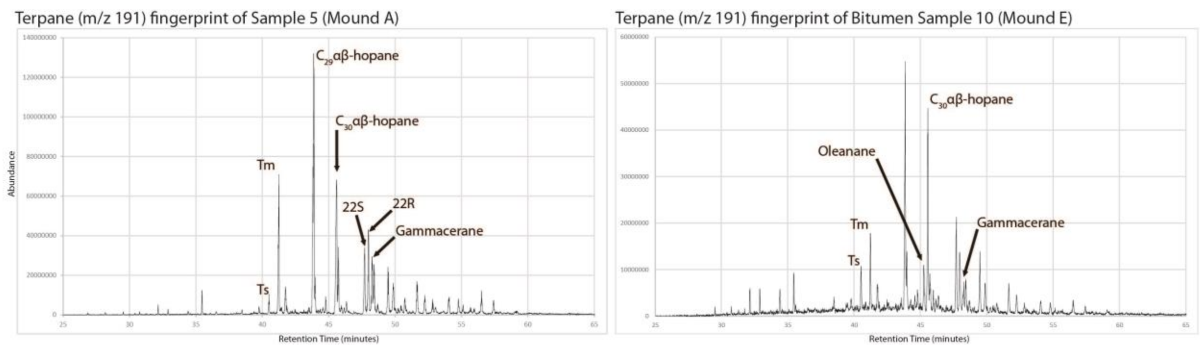

Figure 42 Terpane fingerprints of bitumen Samples 5 and 10. Note the relative low peak of the C_{20}αβ-hopane compound and 22S and 22R (22S-17α(H),21β(H)-Homohopane and 22R-17α(H),21β(H)-Homohopane) in Sample 5, and the presence of chemical oleanane (OLN) in sample 10.

The other 8 samples are reminiscent to many other archaeological bitumen samples and seepages. Figures 40, 41 and 43 document the formation of two clusters; one containing five samples from 'Mackay Tomb 29', 'Mound D' and 'Mound A', whilst the other contains the two bitumen samples from 'Mound N' and one sample from 'Mound A'. Carbon Isotope Analysis (expressed as $\delta^{13}C$) conducted on five of the bitumen samples (with samples from both clusters) gives -27.7 ± 0.1 ‰ and is considered as a too high value for samples coming from the Hit area (with $\delta^{13}C$ of -28.0 ‰ and lower). The measured value of bitumen sample 4 from 'Mound A' (cluster 2) lies in the same range as those from samples from cluster 1 (samples 1, 2, 3 and 8) and does not allow a differentiation based on this parameter. Many of the Iranian seepages can be excluded because of the obvious lack of oleanane in the samples from both cluster 1 and 2.

Molecular ratios and $\delta^{13}C$ values of many archaeological sites have been published in detail by J. Connan in the past, and provide excellent references. It should be noted that the match in molecular ratios is better when considering bitumen from archaeological sites rather than oil seeps, as molecular changes may occur when working source material into bitumen mixtures (Connan and Carter, 2007: 65). Ts/Tm vs. Gammacerane/$C_{30}\alpha\beta$-hopane ratios of bitumen samples several archaeological datasets are plotted in Figure 6. The reference sites used here are several analysed samples from as-Sabiyah (H3), Kosak Shamali, Ra's al-Jinz, and Tall-e Geser (Connan and Nishiaki, 2003, Connan et al., 2005, Connan et al., 2014, Connan, 2010). In the case of Kosak Shamali, only the samples with a northern Iraqi origin have been incorporated here as the other samples have origins which can be excluded for the A'ali dataset.[9] All samples from Tall-e Geser with oleanane present have also been excluded from the data as most of the A'ali samples show no presence of this organic compound. Finally, the samples from as-Sabiyah (H3), an Ubaid-related site have also been incorporated. The main reason for this was the identification of this type of bitumen in an archaeological samples from the Dilmun-period site Tell F6 (Connan and Carter, 2007). The origin of these archaeological samples (H3 and F6) is the Burgan Hill in Kuwait.

Based on the cross-plot of Ts/Tm vs. Gammacerane/$C_{30}\alpha\beta$-hopane (Figure 43) it seems that the bitumen in cluster 1 align best with the samples from the Tall-e Geser (specifically those coming from Sultan), whilst the bitumen from cluster 2 shows close resemblance with several samples from H3 (consequently the Burgan hill in Kuwait). An additional cross-plot (Figure 44) showing $\delta^{13}C$ value vs. Ts/Tm ratio confirms the Sultan-origin for cluster one, but also excludes the Burgan Hill for the cluster 2-bitumen (note that for this cluster, only sample 4 had a $\delta^{13}C$ reading). The apparent higher Ts/Tm- and lower GCRN/$C_{30}\alpha\beta$-hopane of the Cluster 2 samples does however seem to correlate nicely with a set of archaeological samples from Susa (Connan, 2012) and with several of the recently analysed Dilmun-period bitumen from the Kuwaiti-Danish excavations at Tell F6 (Van de Velde, Accepted for Publication-b). Several bitumen samples from both Susa and Tell F6 have been identified as coming from one of the bitumen seepages on the Deh Luran plain.

[9] Other bitumen samples excavated at Kosak Shamali originated form the Hit area in Iraq, and the Samsat seepage in Turkey (Connan & Nishiaki 2003).

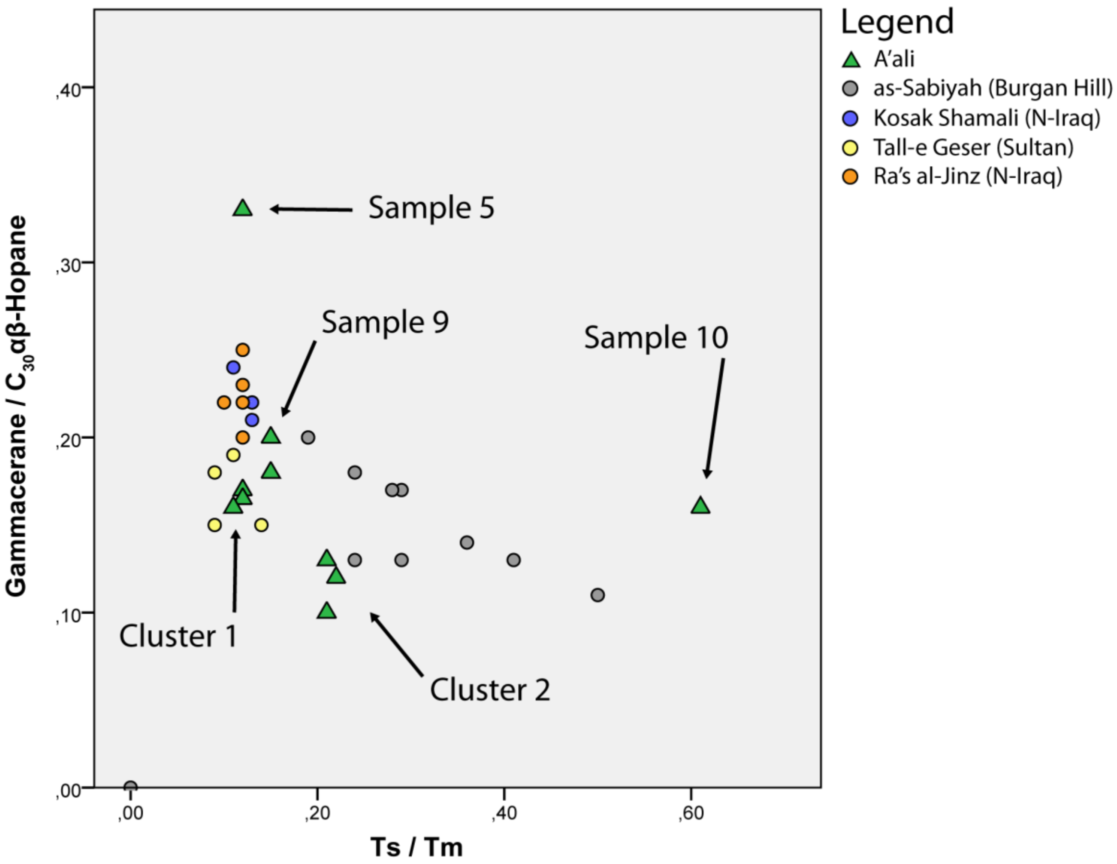

Figure 43 Ts/Tm vs. Gammacerane/C30αβ-hopane cross-plot, including reference samples from as-Sabiyah (H3), Kosak Shamali, Tall-e Geser & Ra's al-Jinz (RJ2).

A remark should be made considering sample 9 ('Mackay Tomb 29'). Statistical analysis (hierarchical- and k-means clustering) favour a northern Iraqi-origin (similar to the Ra's al-Jinz and Kosak Shamali samples) for this sample, rather than Iranian like the rest of the dataset. Considering the fact that this type of bitumen has been attested in Dilmun-related contexts before, this seems a valid option. This, however, remains an uncertainty as the sample might as well simply be a consequence of alteration the sample. A similar northern Iraqi-origin is also possible for Sample 5 ('Mound A'), which, as described above, is considered as an outlier because of the suspected chemical degradation of the sample.

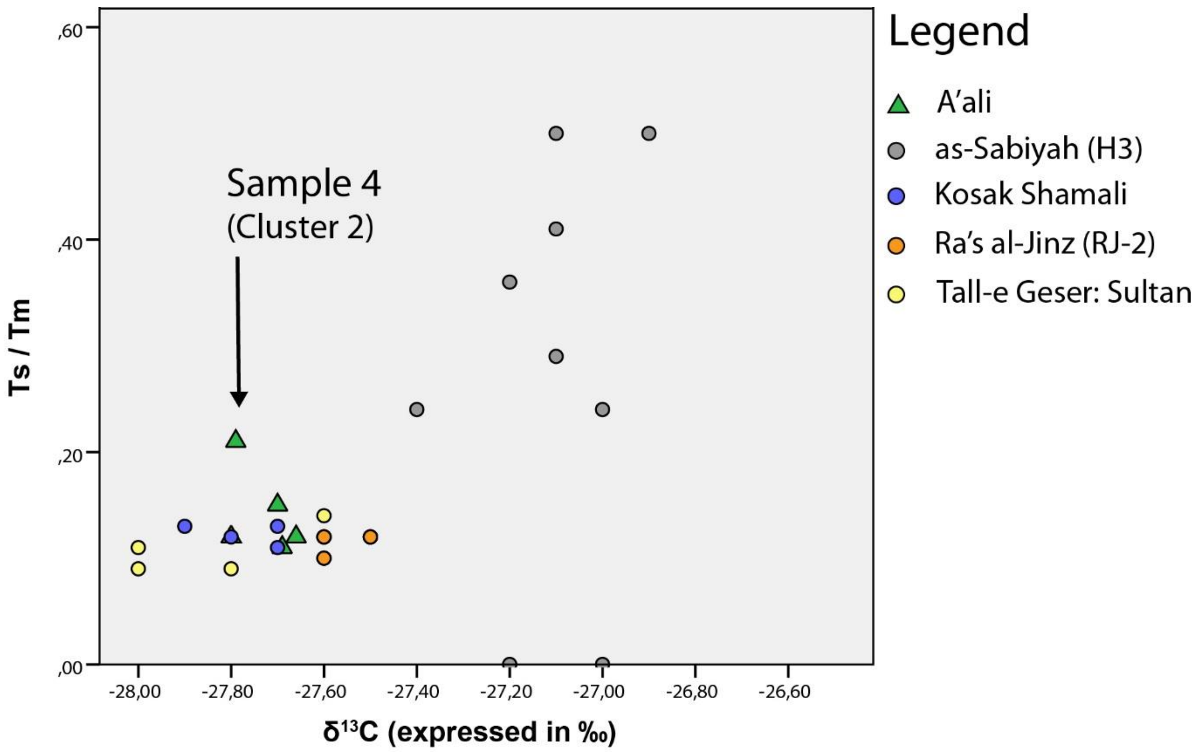

Figure 44 $\delta^{13}C$ vs. Ts/Tm cross-plot, including reference samples from as-Sabiyah (H3), Kosak Shamali, Tall-e Geser & Ra's al-Jinz (RJ2).

Table 7 Measured values for the A'ali bitumen samples.

Sample	$\delta^{13}C$	Ts/Tm	GCRN/$C_{30}\alpha\beta$	OLN/ $C_{30}\alpha\beta$	Identified Origin
Sample 1	-27.8	0.12	0.165		Sultan
Sample 2	-27.7	0.11	0.16		Sultan
Sample 3	-27.7	0.12	0.17		Sultan
Sample 4	-27.8	0.21	0.1		Deh Luran
Sample 5	/	0.12	0.33		unknown/Sultan?/N-Iraq?
Sample 6	/	0.21	0.13		Deh Luran
Sample 7	/	0.22	0.12		Deh Luran
Sample 8	-27.9	0.15	0.18		Sultan
Sample 9	/	0.15	0.2		Sultan?/N-Iraq?
Sample 10	/	0.61	0.16	0.3	Mamatain/Naft Safid

5.2.6 Conclusions

Ten bitumen samples from five different burial mounds from the 'Royal Cemetery of A'ali were selected for geochemical analysis with the aim to determine the geologic origin of the bitumen. Carbon isotope analysis of the asphaltenes fractions and GC-MS analysis on the saturated hydrocarbon fraction were conducted on the archaeological samples, and their results compared with similar data from other archaeological sites and seepages. For the majority of the samples an Iranian origin is most likely, with the bitumen probably coming from the seepage at Sultan (Cluster 1), the Deh Luran plain (Cluster 2), and Mamatain/Naft Safid. Two samples (Bitumen samples 5 & 9) on the other hand are outliers in the dataset, this could be due to alteration of the samples and could have originally come from seepages in Iraq or from Deh Luran , or from a non-identified source.

These results are in line with what was currently known on bitumen from Dilmun. And although bitumen from several seepages was identified, it appears their bitumen was used simultaneously with no chronological distinction..

5.2.7 Acknowledgments

Thomas Van de Velde would like to thank prof. F. Lynen, M. De Vrieze and P. Surmont from the Department of Separation Sciences at Ghent University for their support and help concerning the GC-MS analyses. Also gratitude towards J. Connan for revisions of the data and giving his opinions on the results of the analyses.

5.3 Geochemical analysis on the bitumen lining of a vessel excavated from Tumulus B-5 by Peter B. Cornwall

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Embodying Ancient Dilmun: The Peter B. Cornwall Expedition to Bahrain and Saudi Arabia

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5.3.1 Introduction

This contribution focusses on the chemical analysis conducted on an archaeological sample from Tumulus B-5, the final resting place of a woman whose skeletal evidence has been labelled as individual 12-10146 by Peter B. Cornwall. This individual was buried in a tumulus not far from Qala'at al-Bahrain with an exceptionally large number of objects, one of which an earthenware vessel coated with bitumen on the inside (this vessel has been numbered 9-4700). The purpose of this study is to determine where the bitumen used to coat this vessel was from. As previous studies have pointed out that almost all of the bitumen used in the Gulf are in fact imports from either Iraq or Iran (Connan, 1999a, Connan, 2010, Connan, 2011, Connan and Carter, 2007, Connan et al., 2005, Connan et al., 1998, Connan and Van de Velde, 2010), it was to be expected that the bitumen found in the tomb of individual 12-10146 was also imported from one of the major bitumen exploitation areas[10].

5.3.2 The archaeological sample

As mentioned above, the bitumen analysed in this research comes from the inside-coating of a ceramic vessel (see Figure 45). This is not an uncommon practice, and has been attested at sites such as Qala'at al-Bahrain and Umm an-Namel. A set of 5 bitumen samples from the latter site has been analysed, of which two samples also came from the lining of pottery. One of these sherds was identified as chain-ridged Barbar ware with a red color, whilst the other was noted to being pink in color (Connan and Carter, 2007:143, 146, 148). This is quite remarkable as bitumen was never noted on the inside of Barbar pottery from Qala'at al-Bahrain, but occasionally applied as a thin coating on the interior of imported Mesopotamian vessels (yet infrequent on smaller vessels and never on bowls) (Højlund and Andersen, 1994: 408). The fact that these vessels had a thin bitumen coating suggests that not bitumen was transported –as that would leave larger crusts– but rather other goods of trade with a high value. It is mainly thought that these products would have been fluids rather than solid goods, as the latter don't have the ability to penetrate the vessel in which it was transported anyway. The problem of course is that these products would have been organic in nature and perishable.

[10] For a complete overview of bitumen seepages, see:

- Connan 2012. *Le bitumen dans l'Antiquité*, Arles: Edition Errances.
- Connan & Van de Velde 2010. An overview of bitumen trade in the Near East from the Neolithic (c.8000 BC) to the early Islamic period. *Arabian Archaeology and Epigraphy*, 21, 1-19

Figure 45 Sherd of vessel 9-4700 with the piece of bitumen used for analysis scraped off.

5.3.3 Sample procedures

GC-MS analysis on the saturated hydrocarbons was conducted in order to determine the origin of the sample. The same analytical procedure was used as described in chapter 3.4.

5.3.4 Results

The sample has been measured twice in order to check the consistency of the analysis. No discrepancy between the two measurements was noted. It is the m/z 191 fingerprint (terpane) which holds the most information for fingerprinting the bitumen. Absence- or presence of specific molecules is important, as are quantity-ratios of several specific molecules. The most commonly used molecular ratios are 18α(H)-22,29,30-trisnorneohopane(Ts) to 17α(H)-22,29,30-trisnorhopane (Tm), and Gammacerane (GCRN) to 17 21 -hopane (C_{30} -hopane).

The chromatogram (see Figure 46) shows a molecular pattern quite reminiscent of several bitumen seepages located in present-day Iraq and Iran. The absence of 18 (H)-oleanane in the sample rules out any seepage linked to the Padbeh source-rock formation in southwest Iran (Connan, 2012: 116, Peters et al., 2005b). Remarkable is the low peak of C_{30}αB-hopane as compared to the C_{30}βα-hopane & C_{29}αB-hopane compounds. This type of molecular behaviour is reminiscent of those of two published fingerprints from Ali Kosh and Tepe Tula'i (Iran, Late Neolithic Period) (Gregg et al., 2007), and the molecular ratios commonly used for fingerprinting archaeological bitumen (Ts/Tm and Gammacerane/C_{30}αβ-hopane) correspond more or less with bitumen from the Mamatain seepage in Iran. Bitumen from this seepage however always shows a clear presence of the 18α-oleanane compound, a source-specific marker for Late Cretaceous- or younger rock formations, which is clearly lacking in the sample from the Peter Cornwall collection.

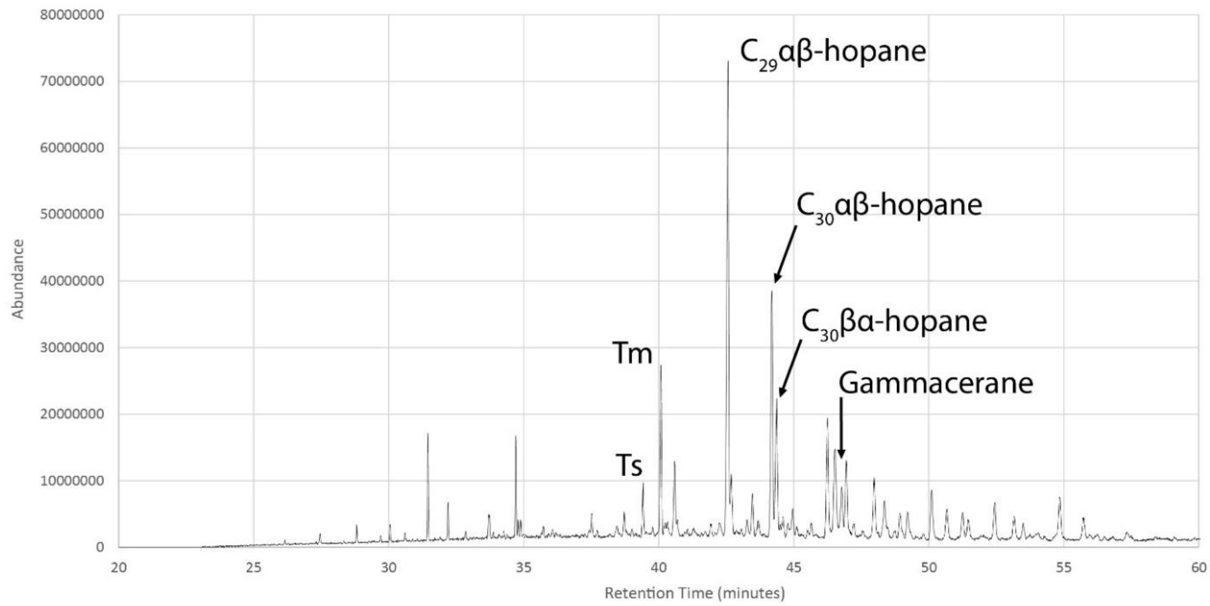

Figure 46 Terpane fingerprint (m/z 191) of the bitumen sample

The Ts/Tm vs Gammacerane/C_{30} -hopane cross-plot (Figure 47) shows molecular ratios from bitumen from various Bronze Age archaeological sites are shown. These sites were selected not only because of their dating, but mainly because their bitumen represent a specific source of origin. In the case of Ra's al-Jinz (dated 2500-2100 B.C.) the bitumen came from the seepages in Northern Iraq, likewise with the bitumen from Umm an-Namel (2100-1000 B.C.) (Connan and Carter, 2007, Connan et al., 2005). The bitumen from as-Sabiyah have been identified as coming from the Burgan Hill seepage (Kuwait), as is the case for one sample from Umm an-Namel. All the other bitumen from the latter site was extracted from the bitumen seepages in the Hit-area in Iraq (Connan, 2010, Connan et al., 2005, Connan and Carter, 2007). It should be noted that the match in molecular ratios is better when considering bitumen from archaeological sites rather than oil seeps as molecular changes may occur when working source material into bitumen mixtures (Connan and Carter, 2007: 65).

The cross-plot (Figure 47) shows that the bitumen sample from the Cornwall-collection doesn't really correspond with any other sample. Also clear is that the bitumen from various sites do not always nicely cluster together. We should always bear in mind that archaeological samples may have been subject to (severe) degradation, causing the alteration of the αβ-hopanes leading to a (sometimes radical) change in molecular ratios such as Ts/Tm and Gammacerane/C_{30}-hopane (Connan, 2012: 129). That is probably also what happened with the sample here at hand, making fingerprinting of this sample difficult.

Considering all factors, we suspect it likely that this sample originally came from Iraq, but was in a high state of degradation and therefore failing to provide a more detailed place of geological origin. The sample could either be coming from the seepages in the Hit, or from those in northern Iraq. Of those 2 options, the former seems the most likely one.

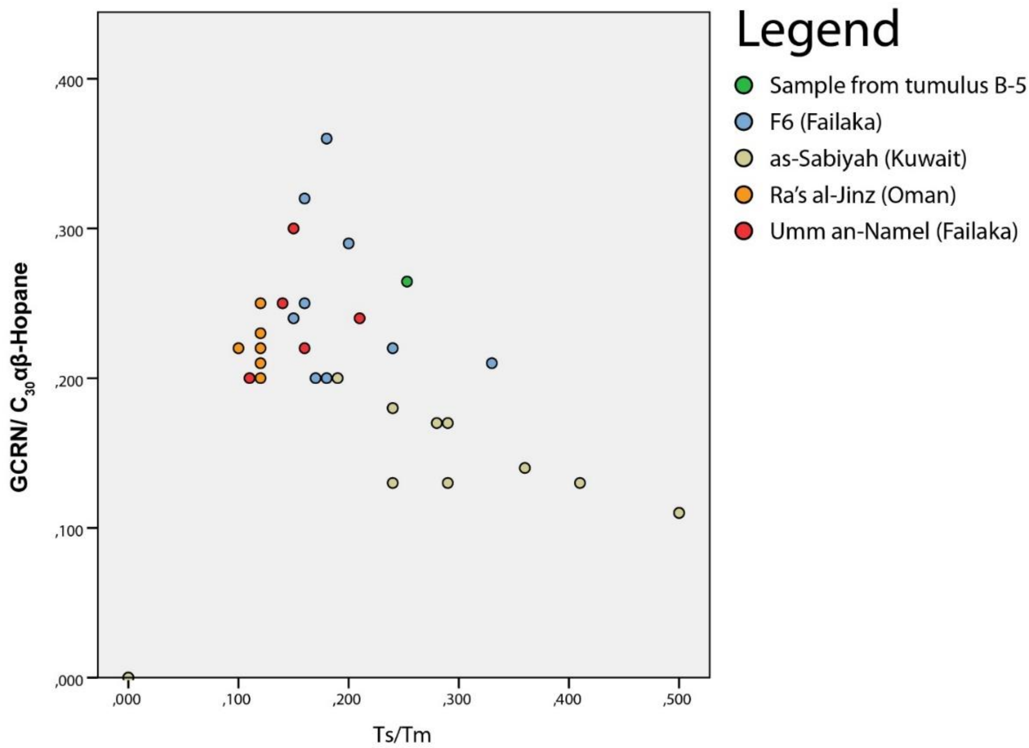

Figure 47 Cross-plot of molecular ratios TS/Tm to Gammacerane/C_{30}αβ-hopane. Molecular values of references retrieved from Connan et. al. 2005; Connan and Carter 2007.

5.3.5 Bitumen in Dilmun

The sample from Tumulus B-5 isn't the only bitumen that was analyzed from Dilmun-contexts, by which we here refer to the island of Bahrain, Failaka, and the Kuwait Bay. Previous studies on contemporary samples have focused on bitumen from Qala'at al-Bahrain, Buri, Karranah, Saar (Connan et al., 1998), F6 (Failaka), Umm an-Namel (Connan and Carter, 2007, Van de Velde, Accepted for Publication-b) and A'ali (Van de Velde and Bodé, Accepted for Publication)

The analysed samples from Qala'at al-Bahrain may further be divided into two different periods: 2500-2000 BC and ca. 1700 BC. All samples have an Iraqi origin, of which the 5 prior to 2000 BC can probably be assigned to the seepages in Northern Iraq (Connan and Carter, 2007: 175). There is unfortunately no further specification available for the 3 samples in the 2000-1700 BC timespan. These findings are in line with the samples from Umm an-Namel (dated around 2100-2000 BC), all 5 of which have been identified as bitumen from seepages in Northern Iraq (Connan and Carter, 2007: 173). Although we must mention that the available data on this excavation is very scant, as the site lacks a comprehensive excavation report.

The usage of bitumen from Northern Iraq in the Gulf is a continuation as this bitumen is already protruding the Gulf from the Late Neolithic Period. The first evidence of this bitumen in the Lower Seas is provided from Dosariyah (Van de Velde et al.) and continued at sites such as Ain As-Sayh site C (McClure and Al-Shaikh, 1993), Ra's al-Jinz (Connan et al., 2005), and Umm an-Nar (Connan and Carter, 2007). Although sample sizes from Ain as-Sayh and Umm an-Nar are extremely small (one sample each), we can consider those from Ra's al-Jinz and Dosariyah as statistically relevant (subsequently 22 from the former and 15 from the latter site).

But around 2000 BC the dominance of Northern Iraqi bitumen seems to come to a halt, as seen in samples from both Failaka and Bahrain. The French excavations unearthed a tower-like structure at F6 (Failaka, Kuwait) (Calvet and Gachet, 1990), of which 10 samples (dated around 2000–1700 BC) indicated a usage of mainly bitumen from the Hit area[11] (Connan and Carter, 2007: 148, 173). Recent excavations at F6 on Failaka by a joint Kuwaiti-Danish team also uncovered many bitumen pieces which have also been subjected to analysis. Preliminary analysis of this bitumen indicates that this bitumen was imported from southwest Iran.

As mentioned above, the early 2nd millennium samples from Qala'at al-Bahrain are said to have come from Iraq, without further specification to either the Hit area or the seepages in northern Iraq (Connan et al., 1998, Connan and Carter, 2007: 175). Remarkably, the bitumen excavated at the settlement site of Saar were all exclusively imported from seepages in southwest Iran (Connan et al., 1998: 170). This bitumen has been found in 7 different buildings spread over the entire site (Killick and Moon, 2005a, Connan et al., 1998), indicating that what we see in the samples is an accurate reflection for the entire site. It is remarkable also Iranian bitumen that has been attested in tumuli from Buri (n=1) and Karranah (n=3) (Connan et al., 1998), but also from several burial mounds from the burial field of A'ali (Van de Velde and Bodé, Accepted for Publication). We should keep in mind that bitumen was not 'used' on burial sites, but rather deposited in the form of grave-goods by people from a (nearby) settlement.

[11] Of the 10 samples analyzed, 9 out of 10 showed a Hit origin (Iraq), while one sample came from the Burgan Hill (Kuwait), a small seepages which was already exploited by the inhabitants of as-Sabiyah at the end of the 6th millennium.

It remains however unclear as to why Saar seems to be supplied exclusively by Iranian bitumen, whilst only Iraqi bitumen was identified at Qala'at al-Bahrain. Possible explanations for this fact might be attributed to the different natures in these 2 settlements: Qala'at al-Bahrain was the main port-of-trade of Dilmun with strong ties to Mesopotamia making it able to easily import materials in large quantities, whilst Saar didn't have these ties and had to rely upon other trading partners (Connan et al., 1998: 176) (Connan and Van de Velde, 2010: 14). Unfortunately the material from Saar doesn't really imply such a condition. The pottery for example, shows an extremely low number of imports, which is in stark contrast with Qala'at al-Bahrain, reflecting Saar's rural character (Carter, 2005a: 266). Also, nothing in the material culture seems to favour Iran as a supplier of foreign materials. It is thus very unlikely Saar functioned as a hub of trade beside the existing metropolis of Qala'at al-Bahrain, its more inland position also isn't very favourable to fulfil that role. Nevertheless the question remains, why the difference in bitumen supplier? Both Saar and Qala'at al-Bahrain are located on the northern part of the island, and it would seem logical that the inhabitants from Saar got their hands on foreign materials through the capital.

On the other hand, the results from geochemical screening of bitumen samples from Early Dilmun contexts at Failaka indicate that bitumen with both Iraqi- and Iranian origin was used at the island. That of course reflects what we see on Bahrain, and we may assume that the bitumen supply was inherent to a certain chaîne opératoire linking the islands Failaka and Bahrain. But if that was the case, we could also expect that the bitumen at Qala'at al-Bahrain and Saar would be more heterogeneous.

5.3.6 Conclusions

The Dilmun Bioarchaeology Project studies the Cornwall-collection, currently housed in the Phoebe A. Hearst Museum of Anthropology at the University of California (Berkeley). This collection includes the contents of Tumulus B-5, a Dilmun culture assigned burial several kilometres southeast of Qala'at al-Bahrain. One of the grave goods (no. 9-4700) was a vessel lined with bitumen to seal the recipient and hold (probably) precious liquids. A small piece of this bitumen was used to conduct geochemical analyses in order to provide an origin for this sample. GC-MS was used to investigate the saturated hydrocarbon fraction and to retrieve specific molecular ratios. Although this process was executed without difficulties, degradation of the bitumen caused the molecular pattern to alter making it impossible to provide a positive match either with seepage-specific data, or with molecular data from bitumen excavated at archaeological sites. All parameters considered, we suggest that this bitumen was imported from Iraq, but it was impossible to determine that this sample came from either the Hit area, or from northern Iraq.

Chapter 6 Hellenistic period bitumen in the Gulf

6.1 Introduction

This chapter discusses the analyses that were conducted on several bitumen samples, all dating to the Hellenistic period[12] in the Gulf. In the advent of my bitumen research, I required a bulk sample of bitumen to be able to experiment with sample preparation of samples and with analytical parameters. As Ghent University was then involved in excavations at the site of Mleiha (Sharjah, United Arab Emirates) where bitumen was found, this formed the ideal starting point for chemical analyses. Somewhat the same goes for ed-Dur, as Ghent University excavated this site from 1987 to 1995 and several bitumen samples were readily available from our depot. Because of the close collaboration between this University and the Emirate of Sharjah, it was no problem to have a bitumen sample from the contemporary site of Dibba sent to Ghent for geological screening. Results of these analyses were presented at the 2013 Seminar for Arabian Studies (London), but have not yet been published.

6.2 Archaeological samples

All bitumen discussed in this chapter come from three Pre-Islamic sites: Dibba, ed-Dur and Mleiha. These sites were selected specifically because they are all situated in the

[12] The term Hellenistic Period is here defined as the range between the third century B.C. and the third century A.D. and not specifically linked to nor the Hellenistic- nor Parthian cultural complex.

same context, the Pré-Islamique Recent C Period (1^{st}-2^{nd} century A.D.) in the southeast Arabian Peninsula.

At the site of Dibba, bitumen from a rather unique context was analysed. Several large storage vessels, completely filled with bitumen, were recovered from a settlement context (Rutten K., pers. comm.). Before this find, it has always been a question whether bitumen was made into a mixture right after collection at the seepage, or transported in its natural from and mixed with other materials at the place of application. This find at least hints at option one for the period of our concern (Connan and Van de Velde, 2010: 15). Several fragments from the content of one of these vessels was used for analysis.

At the site of ed-Dur, bitumen was commonly found as lining of pottery vessels. Three sherds with adequate amounts of bitumen on them were selected, and their lining was scraped off and prepared for analyses. All of these sherds are similar or identical (in fabric and shape) to vessels identified as having a southern-Mesopotamian origin (Rutten, 2006). All three of these sherds were unearthed from settlement contexts.

During the 2009 Ghent University excavations at Mleiha, the remnants of a bitumen basket was unearthed from a domestic context (with possible small-scale industrial activities) dating to the PIR C period (Pincé et al.). Bitumen-coated vessels and baskets are found frequently at archaeological site, especially at Bronze Age sites alongside the shoreline of the Persian Gulf. The woven part; made from palm-leaves or fiber-material, functioned as a frame and was covered on inside and outside with bitumen, leaving a strong and waterproof object. And although the woven part of these objects disintegrate, and the bitumen more than often crumbles and is destroyed, several sites provide us with the bitumen still (partly or even completely) intact. In the case of Mleiha, a complete basket was found in-situ and provided plenty of bitumen for analysis.

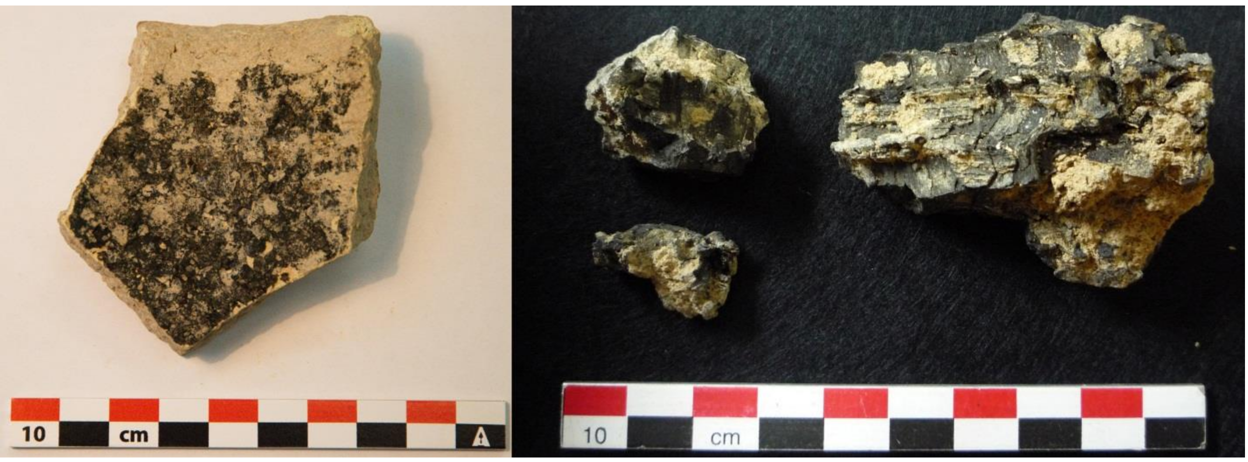

Figure 48 Photo of the coating of one of the ed-Dur sherds used for analyses (left), and some bitumen lumps of the basket that was excavated in Mleiha (right).

6.3 Analytical results

The methodology as discussed in chapter 3.4 was used and will not be repeated here.

The m/z 191 chromatograms from all samples show a strong presence of hopanes and are all reminiscent of the fingerprint of most of the main bitumen extraction sites in the Near East. Remarkable however is the presence of 18 (H)-oleanane in the samples from both Dibba and Mleiha. This compound is an unique marker and occurs mainly in Late Cretaceous or younger rock formations, such as the Padbeh source-rock formation in southwest Iran (Connan, 2012: 116, Peters et al., 2005b). In the case of Dibba, a sample from the same context has been analysed in the past by J. Connan (Connan unpubl.) who also identified chemical oleanane in his sample. A possible source area could be Masjid-i Suleiman or Mamatain, both well-known bitumen seepages in southwest Iran (present-day and during antiquity) of which bitumen was attested at various archaeological sites in Iran (e.g. Susa and various sites on the Deh Luran plain)(Connan, 2012, Marschner et al., 1978: 110), but also on various sites in the Gulf such as A'ali (Bahrain), Akkaz and Failaka (Kuwait) (Connan, 2011, Van de Velde and Bodé, Accepted for Publication, Van de Velde, Accepted for Publication-b).

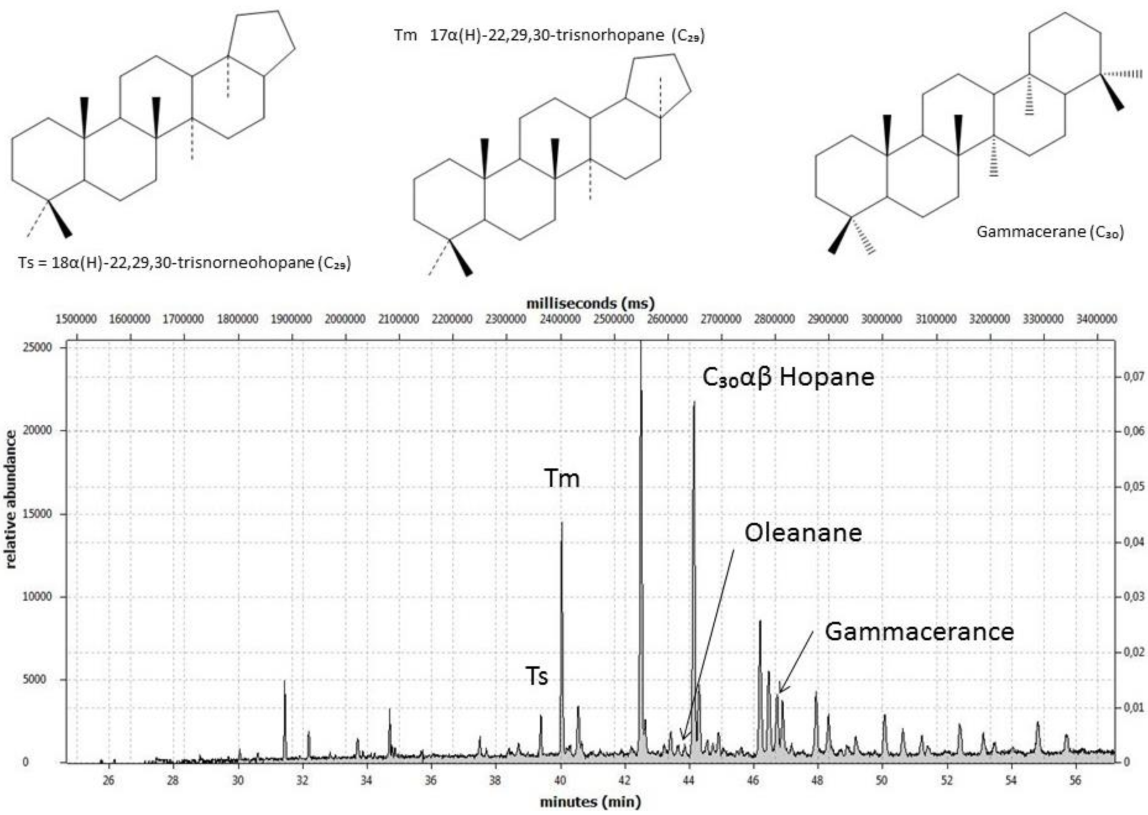

Figure 49 Chromatogram of the m/z 191 fraction of the bitumen sample from Mleiha. Molecules that are later on used in cross-plot for seepage identification are marked (Ts, Tm, Gammacerane, C_{29} hopane, C_{30} -hopane, C_{31} 22R hopane). Structural formulas are also given for several of these molecules.

Cross-plots were made for the most diagnostic compound ratio's, 18α(H)-22,29,30-trisnorneohopane(Ts) / 17α(H)-22,29,30-trisnorhopane (Tm) vs. Gammacerance (GCRN) / 17 21 -hopane (C_{30}) and Ts/Tm vs. Oleanane/ C_{30} in order to further examine the data (See Figure 50). Molecular ratios from other oleanane-containing bitumen samples have been incorporated in these graphs. In general, the oleanane/C_{30} ratio in the samples from Mleiha and Dibba are very low due to the low intensities of the oleanane compound. Such low quantities are not common from any known seepages and were for the first time observed in the archaeological bitumen samples from Akkaz. Although no reference seepage with matching oleanane/C_{30} ratio is known, the Ts/Tm and Gammacerane/C_{30} ratios of the Akkaz-bitumen do match with those from the Deh Luran plain. Considering the fact that the seepages on the latter location do not contain any oleanane, it was proposed that the Akkaz bitumen, in fact, was a mixture from Deh Luran-bitumen and those from another seepage which resources contain oleanane (Connan, 2011). Yet the same molecular ratios are not only observed in the samples here at hand, but also in several samples from Bronze Age Failaka (see chapter 5.1). It is therefore likely that this type of bitumen was extracted from an unknown source rather than being a mixture of several seepages. The contexts from which the bitumen excavated at both Mleiha and Dibba are contemporary to the bitumen from Akkaz, so it is not unlikely that the bitumen found in southeast Arabia passed through other sites such as Akkaz.

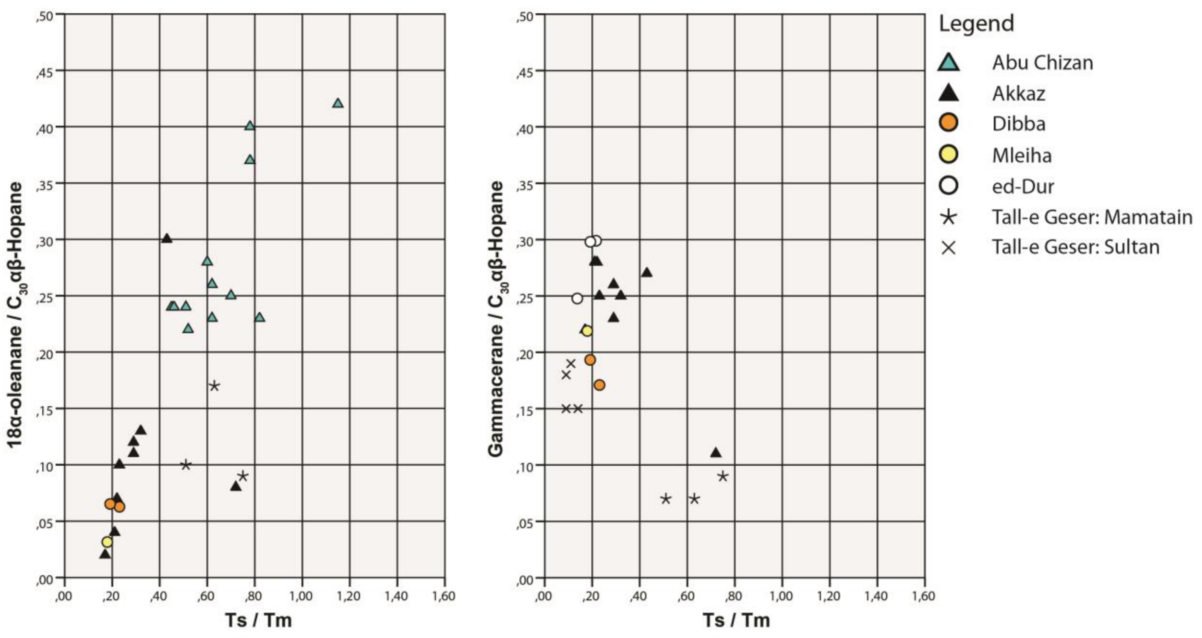

Figure 50 Cross-plots of the diagnostic molecular ratios Ts/Tm vs. 18α-oleanane/C_{30} hopane and Ts/Tm vs. Gammacerane/C_{30}-hopane.

The three bitumen samples from ed-Dur, however, do not contain any oleanane and show a Gammacerane/C_{30}-hopane ratio which is remarkably higher than those from the

Mleiha and Dibba samples. It should be mentioned that this marker is less reliable than Ts/Tm as the $C_{30}\alpha\beta$-hopane compound is more prone to weathering and biodegradation than any other leading to an enrichment of the Gammacerane/$C_{30}\alpha\beta$-hopane ratio. One sample from ed-Dur shows a relative low Ts/Tm ratio (less than 0,2) which is rather typical for bitumen from Sultan (Iran), whereas the Ts/Tm value of the other two samples is rather reminiscent of the bitumen from Hit (Mesopotamia). A higher C_{29} Hopane to Gammacerane ratio in these 2 samples seems to support this possibility. This could indicate that bitumen at the site of ed-Dur was imported from at least 2 different source areas. A similar conclusion was drawn by Jacques Connan after the analyses of several other bitumen samples from ed-Dur, a couple of years ago (Connan unpubl.).

6.4 The PIR-period bitumen contextualized

The Persian Gulf has always been an important sea corridor for trade, already in the Arabian Neolithic Period, as pottery from Mesopotamia was traded in settlements alongside the Arabian shoreline of the Gulf. A tendency that reinforces in later periods. But never has the trade been so intensive and large-scale as during the Parthian Period. The Arabian Peninsula becomes part of larger interaction sphere, spanning from the Roman world to India. This intensification of trade is also visible in bitumen, whereas in previous periods bitumen was a product somewhat limited to coastal sites, it now becomes a very commonly-used material at all sort of sites, also inland sites, such as Mleiha. Enormous amounts of (Mesopotamian) transport vessels coated with bitumen are known from 1st and 2nd century from several archaeological sites; ed-Dur, Mleiha, Larsa, Shimal, Bahrain,... These were undoubtedly used to make the body of the vessels impermeable for safe transport of precious liquids, of which wine is a valuable possibility (Rutten, 2006). Also baskets could be made impermeable like this, as the bitumen from Mleiha prove, but a function in transport seems very unlikely for these objects. They were most likely common everyday objects, as they were during the Bronze Age and even in recent times (Moon, 2005: 196).

6.5 Conclusions

Bitumen samples from three archaeological sites in the U.A.E. were used in this research; being Mleiha, ed-Dur and Dibba. The samples underwent several preparation techniques in order to study the saturated hydrocarbonate fraction, with the ultimate aim to identify the original bitumen seepage of the archaeological samples. The samples from both Dibba and Mleiha contain Oleanane and show a genetic fingerprint similar to that of the bitumen from Akkaz (Kuwait). Although the option was put forward that the bitumen from the latter site was a mixture of oils from different seepages, I would, in light of the findings put forward here, suggest that the bitumen from Mleiha, Dibba and Akkaz all come from an unknown seepage somewhere in southwest Iran. It is suspected that two out of three ed-Dur samples have their origin in Hit (Iraq), whilst the biomarkers of the third sample are more related to the bitumen seepages from Sultan (although a Hit origin remains an option).

The site of Dibba shows that bitumen was not just simply a by-product of trade (unlike the bitumen samples from ed-Dur). Bitumen was intentionally brought into the trading network from southwest Iran, to be used on sites located in Arabia. One of the many applications, could have been the lining of basketry, as our example from Mleiha shows.

A lot of the pottery at the site of ed-Dur was imported material from Mesopotamia, many of them large transport vessels. These vessels were often coated with bitumen to protect the content of the vessels, which were then not the object of trade itself. One can assume that the lining of Mesopotamian pottery vessels was made with Mesopotamian bitumen, which is largely confirmed by the analyses presented here. It implies at least that production of the transport vessels, the bitumen-lining of the vessel, and the filling of the container did not take place at the same time and place. We are situated in a period where a majority of all the goods from- and to the Gulf passes through the Kingdom of Characene, providing a port of trade for not only Mesopotamian- and Gulf-merchants, but also for those from Susiana and southwest Iran. We can presume that Characene was an important market for both Mesopotamian- and Iranian bitumen, not necessarily for own use, but also for large-scale overseas trade.

6.6 Acknowledgments

First of all I would love to thank all the archaeologists who have been supplying me with bitumen samples, making this research possible. For this article in particular, I want to express my gratitude towards Sabah A. Jasim (Directorate of antiquities, Sharjah/UAE) for the sample from Dibba, Ernie Haerinck (Ghent University/Belgium) for letting me use excavated material from the numerous excavation seasons at ed-Dur, and Ernie Haerinck and Bruno Overlaet (Royal Museums of Arts and History, Brussels/Belgium) for the bitumen samples from Mleiha.

Part 3 – From individual datasets to a broader archaeological perspective

Chapter 7 An updated overview of bitumen trade in the Gulf

7.1 The first appearance of bitumen in the Gulf

To date, almost no bitumen indigenous to the Gulf-region has been identified in settlements on the eastern littoral of Arabian Peninsula. The only exceptions being several 6th millennium bitumen samples from H3/as-Sabiyah and one Bronze Age sample from F6, which in both cases were extracted from the Burgan Hill seepage (Kuwait). Generally, the material surfaces convincingly when contacts between the Lower Seas and bitumen-bearing areas (such as Mesopotamia and southwest Iran) start to build up. Contacts between Lower Mesopotamia and the Gulf were established in the Ubaid 3 period and manifested in the appearance of black-on-buff pottery on many Arabian sites. This material, however, is not evenly spread over the many sites dating to the Arabian Neolithic in the Gulf indicating several '*spheres of interaction*' (Carter and Crawford, 2010: 208-209). Although many archaeological sites have been identified and investigated, bitumen was only clearly attested at two sites: H3/as-Sabiyah (Kuwait) and Dosariyah (Saudi-Arabia). Bitumen from the former site has been identified as coming from the Burgan Hill (Kuwait), whereas the latter received its bitumen from northern Mesopotamian seepages (Connan, 2010, Connan et al., 2005, Van de Velde et al., 2015). This difference probably has to do with the dating of the sites; H3/as-Sabiyah (Ubaid 2/3) slightly predates Dosariyah (Ubaid 3) and was inhabited whilst in Mesopotamia still Iranian bitumen was used, as was established for the site of Tell el' Oueili (Connan et al., 1996). In the Ubaid 2/3 period bitumen was probably more scarce than in subsequent periods making it not profitable to redistribute it outside of Mesopotamia —who were importers themselves—, forcing the inhabitants of H3/as-Sabiyah to look for an alternative supplier. During the Ubaid 3 period bitumen seepages in northern Iraq appear to have been more heavily exploited, evidenced by the appearance of this material through the Gulf to Dosariyah (Van de Velde, 2015).

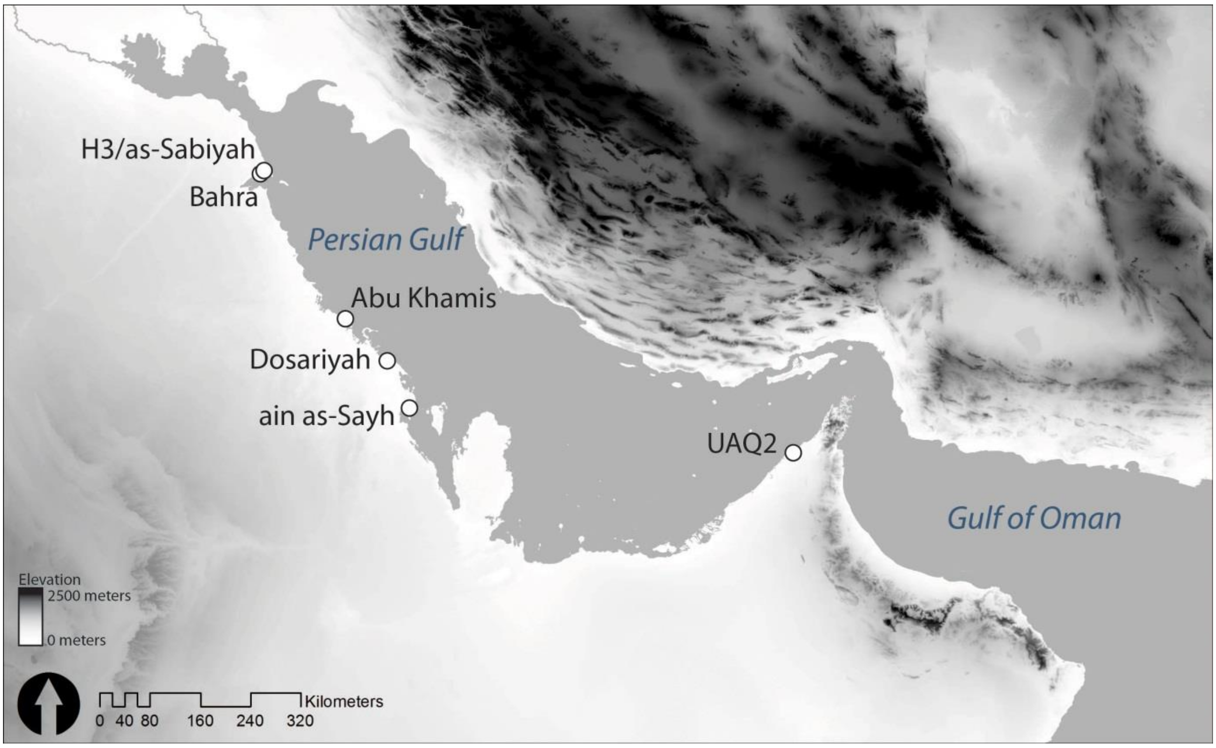

Figure 51 Sites mentioned in this chapter. Bitumen has been attested in considerable quantities at the settlements of as-Sabiyah and Dosariyah. Several bitumen beads have been found in a burial context at UAQ2. Pournelle's (2003) This map is based on Pournelle's reconstruction of the head of the Persian Gulf (Pournelle, 2003).

Bitumen has also been reported from Ain as-Sayh Site C & D in the form of fragments with imprints of woven patterns (possibly related to boats) and as thick crusts on the bottom of earthenware vessels which could be related to bitumen-working (McClure and Al-Shaikh, 1993: 114-122). There are difficulties with dating this bitumen as the site had two occupation phases. The earliest phase dates to the Ubaid 3-4 periods and it was reoccupied during the Jamdat Nasr-Early Dynastic or perhaps even Akkadian interval (Hermansen, 1993: 141). One sample from Site C is said to come from northern Iraq (Connan and Carter, 2007, Connan et al., 2005), but no detailed information on this particular sample has been published.

Several bitumen beads have been excavated from a fifth-millennium burial context in Umm al-Qaiwain, from which the material is said to be coming from Mesopotamia, probably from the seepages at Hit (Phillips, 2002). In contrast to all other bitumen from this period, the bitumen found at UAQ2 was found in a burial context. This may indicate that it was regarded as a luxurious item rather than a utilitarian product. Further, these beads were found on only one individual with several found in-situ around the neck (Phillips, 2002: 176). It therefore appears that we should not regard this bitumen in the same way as that from H3/as-Sabiyah or Dosariyah. In this instance, the necklace —or the beads belonging to it— rather than the raw bitumen material itself, appears to have been the object of trade. The fact that no other bitumen was found on the entire site strengthens this proposal.

It remains striking that bitumen was found in very large quantities at Dosariyah, but practically nowhere else in the Persian Gulf. Although no detailed study on the Dosariyah bitumen has yet been undertaken, it is already clear that the material was used for a variety of applications and was apparently very common. By contrast, bitumen seems to have been relatively scarce at H3/as-Sabiyah. Although this difference may be related to the different dates of these two sites, there is no clear explanation as to why so little fifth millennium bitumen has been reported from other Persian Gulf-sites. Dosariyah is by far the largest site in its timeframe and it possibly functioned as a hub for trade in the rest of the Persian Gulf. As no other sites with bitumen has been identified —with the possible exception of Ain as-Sayh— it would seem that people from Mesopotamia sailed the Persian Gulf as far as Dosariyah, made their intended transactions, and returned. Although Dosariyah is undoubtedly one of the major settlements on the Arabian littoral, this idea seems quite unlikely. Especially considering the large amounts of black-on-buff earthenware that has been identified at Abu Khamis (ca. 83 kilometers north from Dosariyah) and several other minor sites to the south (Masry, 1997, Frifelt, 1989, Matthews, 2001). Remarkably though, no bitumen has been identified at these sites. And although the Ubaid pottery found at Abu Khamis could have been acquired through trade with Dosariyah rather than directly from Mesopotamia, it would seem strange that boats from Mesopotamia passed this site (or in that regard, any other site alongside the Arabian shoreline) without any form of cultural interaction. We should bear in mind that Dosariyah remains the most-extensively investigated and excavated site in this part of the Gulf and bitumen may yet to be found at Abu Khamis and other sites in this area. It has been suggested that Dosariyah was involved in the pearling trade, which seems to have been part of the Neolithic tradition in the Gulf (Charpentier et al., 2012) and probably also the raison d'être for Mesopotamia's initial interest in the region (Oates et al., 1977: 233, Carter, 2005b). Dosariyah is located in the immediate vicinity of pearl beds and is therefore strategically located for either direct or indirect interactions with the inhabitants of Mesopotamia. No pearls have been found at Abu Khamis, yet again, this could simply reflect the smaller scale of excavations.

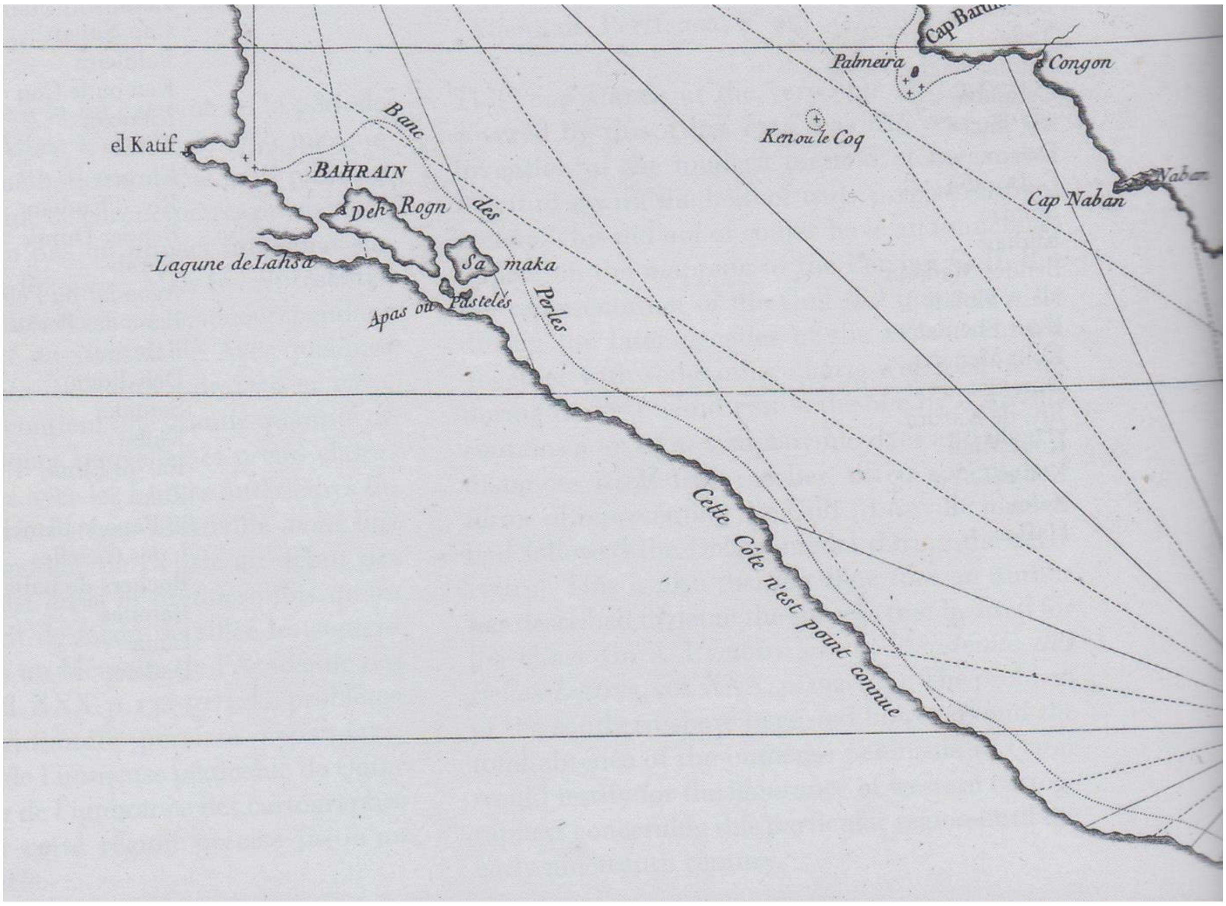

Figure 52 18th Century map (by Jean-Baptiste d'Anville) where the zone containing pearl beds is deliminated (Couto et al., 2006: 294, 296). The zone with pearl beds northern extent reaches up to the city of el Katif, roughly 45 kilometers southwards from Dosariyah.

Our knowledge on the northern part of the eastern Arabian littoral is extremely limited, making it difficult to propose a hypothesis on the nature of the bitumen trade during the Arabian Neolithic, or for interactions on a larger scale. In fact, only three sites have been intensively excavated; Bahra and as-Sabiyah in Kuwait, and Dosariyah in Saudi-Arabia. The former two and the latter also situate themselves in another 'sphere of interaction', a concept which was defined and explained in chapter 4.2. Although down-the-line trade of Ubaid pottery along the shoreline of Arabia seems most likely, it is not impossible that seafarers from Mesopotamia sailed all the way down to Dosariyah to obtain precious goods such as pearls. Quite possibly bitumen found its way to Arabia as part of those endeavours. Whether direct trade or down-the-line trade, it is highly likely that more bitumen is to be found at more sites on the stretch of land between Bahrain and Iraq.

In contrast to the intense settlement-occupation and interregional contacts documented in the fifth millennium, there is practically no information available for the fourth millennium in the Persian Gulf. The only exceptions are the large shell middens found along the Omani coast from Ra's al-Hamra to Ra's al-Hadd and beyond (Uerpmann, 2003: 74), the late fifth millennium site of Akab (Méry et al., 2009), and the site of Al Markh on Bahrain (Roaf, 1974, Roaf, 1976). It has been suggested that the changes in occupation and subsistence strategies in Arabia during this period are linked to climate change (Magee, 2014: 74). The pottery from Al Markh seems Mesopotamian in origin, but for most sites there is little evidence of continuing contacts with Mesopotamia. In addition, no bitumen dating to this period has been identified, with the exception of RH-5 (Oman) where a jar of burnished grey ware, showing indications of bitumen heating, was found in a fourth millennium context (Marcucci et al., 2011: 205). It has however been argued that this vessel may be intrusive (Potts, 1993a: 180). The lack of any other imports at the site supports this argument. To my knowledge, no bitumen dating to the fourth millennium has been reported from any site in Arabia. Indeed, very few imported material in general has been identified at fourth millennium Arabian sites, indicating a decline in relations between Mesopotamia and the Persian Gulf during this period..

7.2 Bitumen in the Bronze Age

7.2.1 Introduction to the Bronze Age and its bitumen

Around 3000 B.C. more archaeological sites and evidence begins to emerge on the Arabian peninsula, especially in the southern part with the appearance of tombs belonging to the Hafit cultural horizon. Only two settlements dating to this period, Hili 8 in the Al Ain oasis (U.A.E.) and HD-6 (Ras Al-Hadd, Oman) (Potts, 1990a: 78, Azzarà, 2009). During this period, imports from Mesopotamia again start to appear in this part of Arabia in the form of pottery found in graves. These ceramics have not yet been identified in the settlement areas, and it has been suggested that in general pottery was poorly integrated into everyday life at the beginning of the third millennium (Cleuziou and Méry, 2000). In any case, their presence is an indication of interaction with inhabitants from Mesopotamian, although it is possible that they ventured only as far as Bahrain (Potts, 1990a: 85-91).

A sudden rise in settlement numbers is documented around 2500 B.C. with the advent of the Umm an-Nar period in southern Arabia, named after the culture which was first identified on the eponymous island at Abu Dhabi. Settlement-intensification dating to the middle of the 3rd millennium has been identified through surface survey in eastern Arabia, with Tarut probably being the most famous site (Crawford, 1998: 38-43). Arabian ties with Mesopotamia seem to grow stronger during this period due to the demand for copper by the latter, thus including Arabia in the Bronze Age World System (Warburton, 2011). The intensification of interregional trade enabled the import of materials into Arabia, such as bitumen. Bitumen is attested at several sites dating to the second half of the third millennium such as Umm an-Nar, RJ2/Ra's al-Jinz, Umm an-Namel and Qala'at al-Bahrain; and was commonly extracted from the seepages in northern Iraq.

This changes around 2000 B.C. when Dilmun becomes the major port-of-trade in the Gulf and the direct contacts between Mesopotamia and Magan cease. Bitumen from then on is omnipresent on both Dilmunite funerary contexts and settlements. Substantial amounts of natural asphalt have been found at Tell F6 and al-Khidr on Failaka, Qala'at al-Bahrain, Saar, Buri, Karranah and A'ali on Bahrain (Belenová-Štolcová, 2010, Calvet and Gachet, 1990, Højlund and Hellmuth Andersen, 1997, Connan et al., 1998, Van de Velde and Bodé, Accepted for Publication, Højlund and Abu-Laban, in publication).

In the following chapters we will take the results from analyses on the Bronze Age bitumen samples and put them in a broader framework of Bronze Age trade in the Persian Gulf.

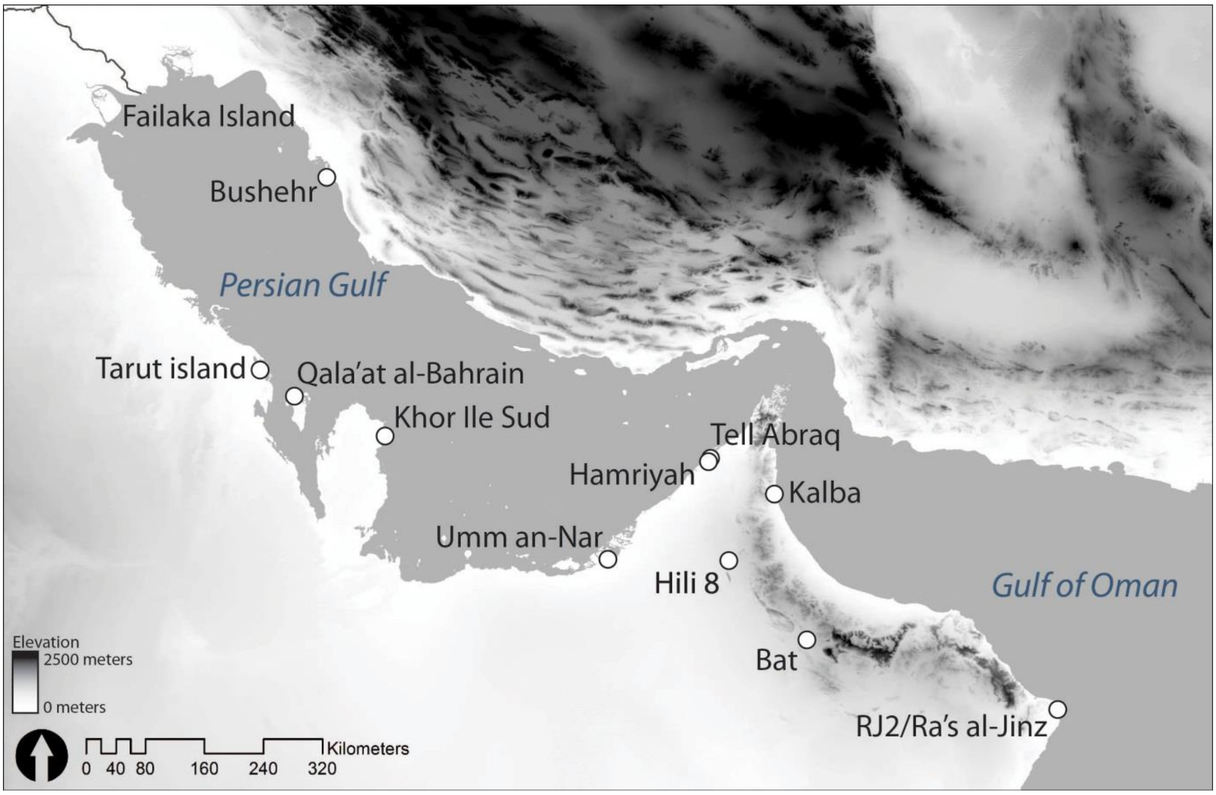

Figure 53 Location of the sites discussed in this chapter. For the sites on Failaka and in the Bay of Kuwait, see Figure 32.

7.2.2 The old data

The analysed datasets from two major Dilmun-culture archaeological sites contribute greatly to our understanding of bitumen trade in this period and context. The results from bitumen analyses of samples prior to this research enabled researchers to form a model of bitumen trade (Connan and Carter, 2007, Connan and Van de Velde, 2010). This model can roughly be split into two major steps with 2000 B.C. as a turning point. Prior to this tipping point almost exclusively bitumen from northern Iraqi sources reach settlements in the Gulf such as at Umm an-Nar (Frifelt, 1995), RJ2/Ra's al-Jinz (Connan et al., 2005), Dosariyah (Van de Velde et al., 2015), Umm an-Namel (Connan and Carter, 2007) & Qala'at al-Bahrain (Connan et al., 1998). This is also in agreement with several cuneiform texts on bitumen in Mesopotamia; the Gudea cylinders (dated to the 22nd century B.C.) narrate the building of the mythical Ningursu temple and explicitly refer to Madga (northern Iraq) as a source of the bitumen (lines 424-433) (The ETCSL Project, 2006) and Ur III period texts clearly state that the provinces of Kimash & Madga, both situated in northern Mesopotamia, deliver huge amounts of bitumen to the capital of the empire, Ur (Connan and Deschesne, 1996: 31). At the turn of the millennium, mainly bitumen from the Hit seepages has been attested in Dilmun-related context at both Tell F6 (Connan and Carter, 2007) and Qala'at al-Bahrain (Connan et al., 1998). Strangely enough, rather Iranian than Mesopotamian bitumen was identified at the settlement of Saar and from several burial tumuli on the island of Bahrain (Connan et al., 1998). Interpretations for this discrepancy were sought in the different natures of the settlements and the different spheres of interaction in which they may have been involved (Connan et al., 1998, Connan and Carter, 2007, Connan and Van de Velde, 2010).

7.2.3 New contributions

New analysis on Dilmun bitumen shows why these hypotheses were not truly convincing, as they point out that Iranian bitumen was in fact not a discrepancy but probably a rather common product alongside Mesopotamian bitumen. Not only was this bitumen (again) identified in several Bahraini burials (Van de Velde and Bodé, Accepted for Publication), but also at Tell F6 on Failaka (Van de Velde, Accepted for Publication-b). This research adds up the total number of bitumen samples from Tell F6 up to 23 (see Figure 54), making the majority of samples of Iranian origin.

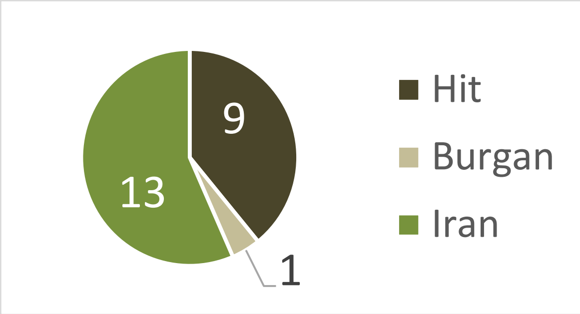

Figure 54 Bitumen samples from Tell F6 on their origins.

The newly-analysed bitumen dataset from Tell F6 was excavated from several trenches in between the temple (excavated by the French missions in the 1980's) and the Palace (excavated by the Danish missions in the 1960's) (see Figure 55). Bitumen from both campaigns have been unearthed at different archaeological levels and contexts and are therefore not a result of a single event.

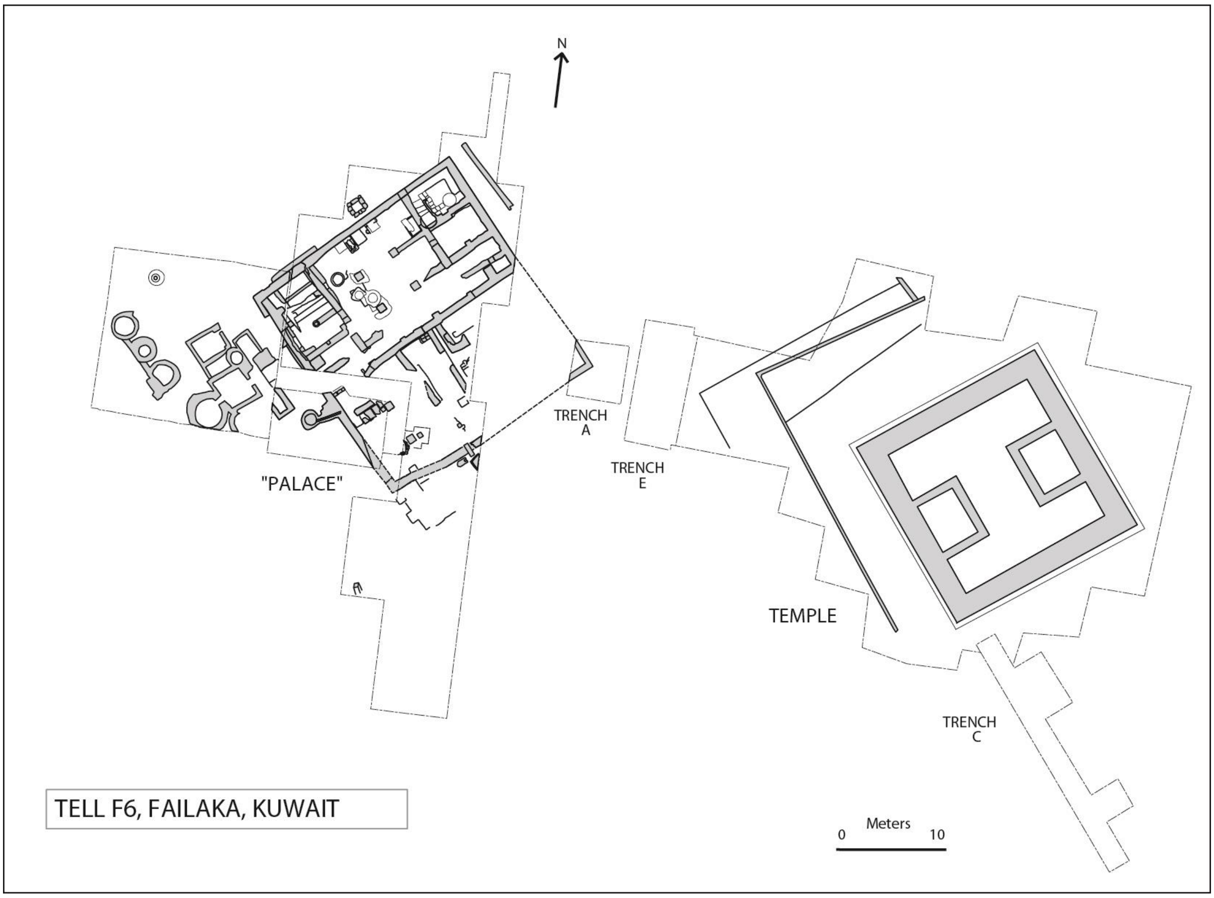

Figure 55 Plan of Tell F6 (Failaka) with trenches from the recent Kuwaiti-Danish mission marked. Image retrieved from (Højlund, 2012).

Prior to this research bitumen from several burial mounds from Bahrain was investigated, but on a very small scale, one sample from Buri and three from Karranah.

All four of them are fragments of bitumen coated baskets, a common grave-good for this period and time, and likely originate from Iranian seepages (Connan et al., 1998, Højlund, 1995). In agreement with these results is the analysis of a new set of bitumen samples which was unearthed from several tumuli belonging to the Royal Mounds at A'ali, which results also point towards southwest Iran as a main source for its bitumen (Van de Velde and Bodé, Accepted for Publication) (see chapter 5.2). This is not surprising as also the bitumen excavated at the settlement site of Saar are Iranian in origin and show strong ties with the bitumen from Tell F6 (see Figure 56). Molecular ratios of samples from Tell F6 and Saar seem to match quite well, although the $\delta^{13}C$ between samples differ slightly. The samples lacking oleanane from both sites are imported from seepages form the Deh Luran plain in southwest Iran, but not coming from the same seepage. The $\delta^{13}C$ values of the Tell F6 samples correlate with those from the Aïn Gir seepage (or perhaps even Deh Luran seepage), whereas those from Saar rather relate to those from the Chersh Merghir seepage (see Figure 18).

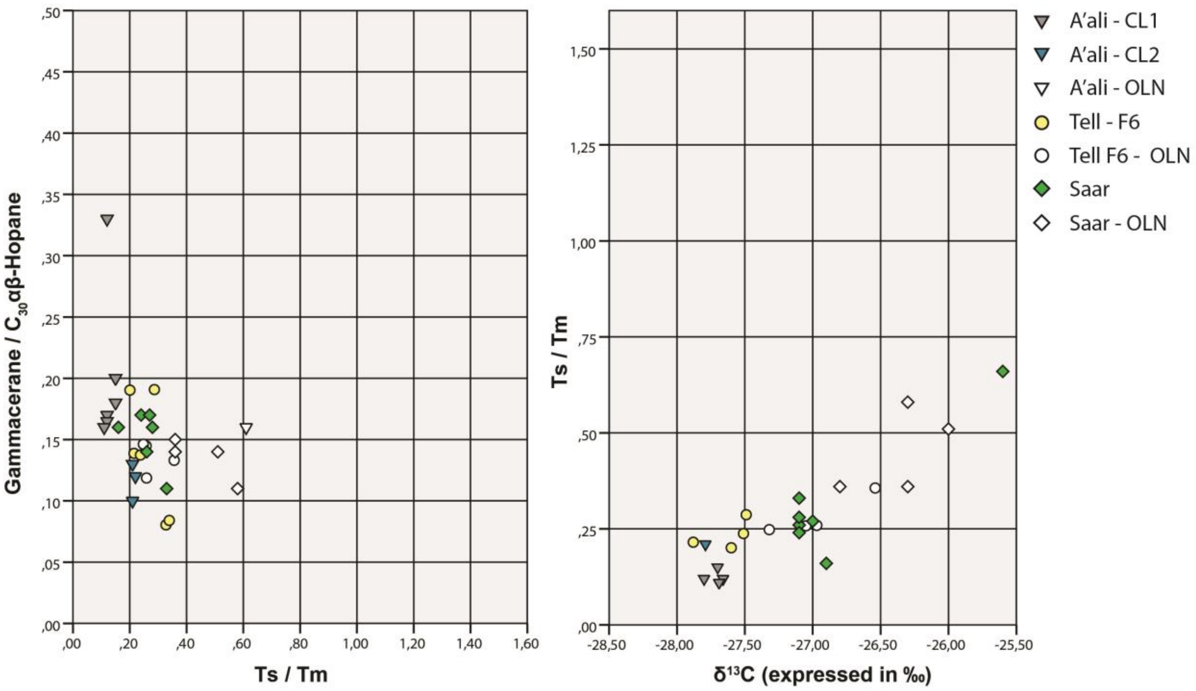

Figure 56 Cross-plot of molecular ratios and $\delta^{13}C$ for the Dilmun archaeological bitumen which have been identified as Iranian in origin. Special thanks to Jacques Connan for supplying me with the raw data concerning the Saar samples.

Based on the results of recent analyses, older models of bitumen trade need to be replaced with a new, more nuanced, model. In particular, the more extensive role of Iranian bitumen in the Gulf should be incorporated, although it remains difficult to assess the ratio between Iraqi- and Iranian bitumen for this period due to the fairly limited sample size from sites. The settlement-sites from which bitumen was analysed, including the number of samples, are shown in Table 8.

In total, the geological origin of 36 samples has been established, of which a majority, 23, originate in southwest Iran. This view is obviously skewed due to the low number of samples from Qala'at al-Bahrain, which is considered to be the major settlement on Bahrain and the 'capital' of the Dilmun geographical area & culture. When taking the burial mounds into consideration, most of its bitumen material has also been identified as Iranian in origin. Considering the common nature of the material and its utilitarian, rather than decorative or luxury function, it should not come as a surprise that the origin of the material found in Dilmun funerary contexts is a reflection of that used in the contemporary settlements. Nevertheless the fact that Iranian bitumen appears to form an important component of bitumen found in the Gulf, perhaps even being the dominant source, does not exclude the presence of Iraqi bitumen at sites such as Tell F6 and Qala'at al-Bahrain.

The apparent difference in source of the bitumen between the samples unearthed at Tell F6 by the French missions from the temple and those by the Kuwaiti-Danish campaigns remains striking. One of the possible explanations is that a shift between suppliers took place at certain time, prior to which Iranian bitumen was used (cf. Saar on Bahrain) and after which Mesopotamian Hit-bitumen was more readily available. The current available data on the samples, however, makes it hard to prove such a hypothesis. As of yet, no detailed information concerning the contexts of the Kuwaiti-Danish samples, nor their exact dating is available (though an extensive excavation report will appear shortly). In addition, it is also possible that the initial dating of the French bitumen samples is not entirely correct and that these samples are more recent and should be dated to the middle of the 2nd millennium B.C. rather than to the beginning of this millennium. The same goes for the three Early-Dilmun samples from Qala'at al-Bahrain: the published records concerning these samples fail to give exact localization of these samples, and quite possible, they are intrusive from the upper layers. Additionally, the number of analysed samples available for this period on Qala'at al-Bahrain is very low (n=3).

In regard to all these problems concerning dating and contextualisation of the Early Dilmun samples from Tell F6 and Qala'at al-Bahrain, there remains little to defend a presence of Hit-bitumen on Dilmun contexts dating to the late 3rd- early 2nd millennium B.C. It would especially explain the problematic difference observed in the bitumen samples from Tell F6.

Concerning the Early-Dilmun period in the Gulf, bitumen from Iranian seepages is not an unlikely idea since ties between Elam's involvement in the Gulf have been attested (Potts, 1999: 178-181). In fact, also bitumen has been found (though not sourced) on the 2nd millennium Elamite settlement on Bushehr (Iran) (Pézard, 1914). It leaves us to wonder if there were exchange networks on the eastern shorelines of the Persian Gulf beside the well-established ones alongside the Arabian littoral. The intensity of the sites on the Bushehr Peninsula (Carter et al., 2006) at least seems to indicate that we may underestimate the role of this coastline in the interregional trade in Antiquity, and possibly bitumen and other products travelled overland to Bushehr prior to getting shipped to Gulf-settlements such as Saar on Bahrain. In any way, the old models on bitumen trade in the Gulf for the Early Dilmun-period seem to grossly underestimate the role of Iranian bitumen.

Table 8 Dilmun-period settlement-sites from which bitumen has been analysed, including the number of bitumen samples.

Site	No. of samples	Iraqi origin	Iranian origin	Burgan Hill	Uncertain origin
Tell F6 (Failaka)	23	9	10	1	3
Saar (Bahrain)	13	0	13		
Qala'at al-Bahrain (Bahrain)	3	3			
Total	39	12	23	1	3

7.2.4 Changing patterns and ideas for 3rd millennium bitumen

After having reviewed the analysed bitumen and the information they provide on ancient trade, it is time we put this all into a more archaeological-economic perspective. After all, the use of bitumen and the change in suppliers are a result of a chain of events and circumstances on many levels: political, social, economic & geophysical.

Chemical analysis (including lead-isotope and compositional analysis) of copper from both Gulf- as Mesopotamian sites have proven the long-standing hypothesis, entailing that the main thriving factor in the Mesopotamian-Gulf trade was the copper ore exploited from the Hajjar mountains (Weeks, 1999, Weeks, 2003, Begemann et al., 2010, Begemann and Schmitt-Strecker, 2009). As Mesopotamia was poor in metals, they looked towards the settlements in the Gulf for obtaining this richness. On the Ur III period, Steinkeller (2004: 103) writes:

> "Since the merchants were institutionally part of the provincial organization [...] they also fall under the administration of the central government" (Steinkeller, 2004: 103)

If prior to 2000 B.C. this trade was organized by one central administration/government as Steinkeller (2004) and Magee (2014: 117) suggest, then also the logistics of these operations would be handled by the same institution. Indeed, cuneiform texts from the Ur temple accounts often mention the enormous "*Tilmun-boats*" (Oppenheim, 1954), which were an essential factor in maintaining the maritime network. Whether these boats were initially fabricated from either wood or reed bundles, no matter what bitumen always was a vital material in the construction for which massive quantities were present (Potts, 1995, Carter, 2012). This brings us again to Hammurabi's wish to annex Mari because of its bitumen and the possibilities and economic advantages that the natural resource offered (cfr. Chapter 2.3.1).

Several studies seem to prove that the boats active in the Gulf and used to obtain the precious copper from Magan, were Mesopotamian boats. By Zarins (2008), for example, who acknowledges this based on the corpus of cuneiform tablets from Umma handling on dockyards and the construction of **má-má-gan** and **má-dilmun** ships. This is backed up by the bitumen finds from Ra's al-Jinz, which bear bulrush reed impressions, a species indigenous to Mesopotamia and not growing in Oman (Cleuziou and Tosi, 1994: 754).

I think that we have to explain the presence of bitumen in the Gulf during the late 3rd millennium in exactly this regard: not as an everyday commodity or product, but specifically intended for the maintenance and repairing of Mesopotamian ships and consequently the upkeep of the entire trade network. Travelling from south-Mesopotamia down into the Gulf is not without any peril, and we cannot accept that every ship made the travel without any form of havoc. Hulls of ships could easily be repaired if bitumen was beforehand (see the anthropological reference in chapter 2.3.3.2), and as there were no active seepages on the Arabian Peninsula in Antiquity, this material must have been supplied by Mesopotamia. Then either bitumen was deliberately dropped off at Gulf-settlements by Mesopotamian ships in order to supply building material(s) to anticipate future repairs for another member of the convoy (or rather 'trader'), or ships were stripped from their bitumen by the inhabitants of Gulf-sites and stored for later use (either for Mesopotamian ships, or for their own). Considering the fact that the boats themselves were Mesopotamian-built, it is logical to assume that their maintenance was also taken care of by its makers.

One of the aspects that led me to this theory was the scarcity of bitumen on many sites, and if it was present, its location on the site. As mentioned in Chapter 2.3.2, bitumen was often found in one specific building or room on the site, functioning as a warehouse or location of storage. Bitumen attested at these types of contexts often also had marks on them testifying of a previous usage, which in turn indicates that the material was valuable, scarce, and that it was preserved for specific functions. Although not all contemporary, bitumen was attested in this manner at H3/as-Sabiyah, Ain as-Sayh Site Site C, Umm an-Nar & Ra's al-Jinz (Carter, 2010, McClure and Al-Shaikh, 1993, Frifelt, 1995, Cleuziou and Tosi, 2000). Especially the sites of Ra's al-Jinz and Umm an-Nar Settlement are interesting here, as these sites are contemporary (dating to the second half of the 3rd millennium) and were main hubs in the Mesopotamian-Magan trade of the late 3rd millennium B.C. Umm an-Nar island is located strategically with regards to the maritime trade with Mesopotamia, whereas Ra's al-Jinz seems to have had strong ties with Meluhha (Magee, 2014, Frifelt, 1995, Cleuziou and Tosi, 2000). Many of the bitumen found at the latter site show impressions of reed and ropes, but also barnacles are observed in many samples, showing that this bitumen was used in naval architecture before and that these pieces came from stripped hulls (Connan et al., 2005). Little Mesopotamian imports were noted from RJ2, with the exception of about twenty body sherds from large buff ware. Remarkably all of these fragments show traces of bitumen, on the inside but sometimes also on the outside, suggesting that these sherds were part of vessels used to transport bitumen (Cleuziou and Tosi, 2000: 53). These findings support the hypothesis constructed above and indicate that the supply of bitumen came directly from Mesopotamia rather than simply from the stripping of ships. The problem however, is that there is scarcely any bitumen reported from Magan-period settlements in the Gulf. Although many Magan sites have been identified and a handful excavated, bitumen seems to have been limited to the two sites mentioned above. And although bitumen has been found in mid-2nd millennium contexts from Tell Abraq (Magee, pers. comm.), none was identified in 3rd millennium strata, which is quite remarkable considering the vast number of imported materials that was excavated from this site (Tengberg and Potts, 1999, Weeks, 1997, Potts, 2000). One possibility is that the excavators just haven’t touched upon the contexts where bitumen is to be found, which is possible when bitumen was kept and stored on one specific location. Another possibility would be that Tell Abraq didn’t belong to the coastal network in which Mesopotamians ventured. This could potentially be backed up by the fact that no pottery older than the Old-Babylonian Period was identified at the site (Potts, 1991), but as we identify Ra’s al-Jinz as a node in the Mesopotamian-organized network based upon a handful of sherds, it just doesn’t seem fair to deny Tell Abraq to this role solely on the lack of some sherds, which may actually be present on the site but just not yet unearthed. Tell Abraq, however, isn’t the only site which lacks bitumen in this period. In fact, there are very little archaeological sites from which bitumen was reported, the only exceptions being the ones we’ve already mentioned. For a comprehensive overview of all reported sites of this period, see Magee (2014). The lack of bitumen on any inland site and from the burials again indicates the scarcity of the material for this area in this particular period. The strictly coastal nature of bitumen on the other hand hints to a close relation with either the trade networks, the actual boats, or both. Finally, considering the strong institutional power and centralized government of the Ur III empire, it may not come as a surprise that the marine trade was supported by strong logistics.

7.2.5 Second-millennium changes in bitumen and economy

Things however were bound to change, and at the turn of the 3rd to the 2nd millennium all factors were present not only for an extensive reshaping of the Mesopotamian-Gulf trade contacts, but also of the Arabian eastern littoral itself.

Højlund (2008: 136) notices that from Qala'at al-Bahrain Period II onwards the Early Dilmun Society becomes more wealthy and expands dramatically. He relates this to an intensification of trade through the Gulf caused by a Mesopotamian increased demand for foreign commodities and raw materials; which was consequently monopolized by Dilmun. The changing political situation in Mesopotamia most likely was the thriving force behind this, and Laursen (2009) suggests that the conflict between the Ur III state and Anshan and Shimashki may have contributed greatly to the demise of Magan in this period. With the raging conflict between the two states, southern Mesopotamia may have ceased its longstanding contacts with the rest of Iran and particularly Marhashi, which in his turn was Umm an-Nar's ally or possibly even its hegemon (Laursen, 2009). These diplomatic issues may have heralded the end of the economic transactions between states and creating a vacuum from which Dilmun took advantage (Højlund, 2008: 154, Steinkeller, 2006).

It is generally accepted that the main thriving force behind this trade remained copper, with the only difference that it is now the Dilmun-traders who supply it to Mesopotamia, rather than Mesopotamians obtaining it themselves in Arabia. With the establishment of a port-of-trade on the island of Failaka, Dilmun had everything in it to control the Gulf-trade and enrich itself, which is exactly what happened around 2000 B.C. Olijdam (2014) even suggests that the rise of Dilmun was not a fortunate chain of events, but a rather planned strategy in which Dilmun dictated the rules of the game.

A curious observation is the demise in number of Mesopotamian-imported ceramics at Qala'at al-Bahrain at time of the Isin-Larsa period in comparison to the previously era (Højlund and Andersen, 1994). Carter (2003: 37) looks at Failaka and its establishment as Dilmun's main port of trade in the Northern Gulf as the main reason for this. Earlier colonization of the island of Failaka was impossible as it was submerged until about 2000 B.C. (Lambeck, 1996). Carter's hypothesis probably intertwines closely with that of Laursen (2011), who rather explains lowering number of imported Mesopotamian pottery at Qala'at al-Bahrain by the changing nature of the Mesopotamian trade at that time, i.e. the change from large-scale Ur III enterprises of the Magan-trade towards a more contracted network. On this changing character of the Mesopotamian-Dilmun trade during the Isin-Larsa period Crawford (2005: 44) notes:

> "By the time of the famous Ea-Nasir of Ur, in the Isin-Larsa period, it is clear that private capital was playing an important part in the Dilmun trade. [...] They seem, at least in the case of the merchants, to have been organized into houses or guilds of some sort with officials who acted on behalf of the whole group."

And although the main product of the entire Mesopotamian-Dilmun economy remained unchanged, the by-products of the trade changed. Which is also observable in the bitumen record. Whereas bitumen in the previous period was probably a means to support the trading network, it now becomes an actual product of trade. And although now suddenly massive amounts of this material are to be found in the upper part of the Gulf, it is never attested at sites that are not belonging to the Dilmun-culture complex. On the sites, however, that are Dilmun in nature bitumen is found in large quantities and used for a wide array of applications. Remarkable is the fact that bitumen is now very frequently attested in the burials, not only in the construction of the monuments, but also in the form of bitumen-covered baskets and beakers as funerary gift (Højlund, 1995, Daems and Haerinck, 2001, Connan et al., 1998). As bitumen artefacts were prior to 2000 B.C. almost strictly found on settlement-sites in the Gulf, their sudden and omnipresent appearance on Dilmun burial sites is noteworthy and definitely attests to a sudden larger availability of the resource itself. As the bitumen in this period is —just as in earlier periods— imported, it attests to changing and more intense trading networks with both Mesopotamia and southwest Iran (Connan and Carter, 2007, Van de Velde, Accepted for Publication-b, Connan et al., 1998, Van de Velde and Bodé, Accepted for Publication). The increase in the usage and trade of bitumen is a result of the changing economic situation orchestrated by Dilmun in the early 2nd millennium B.C. But bitumen is just one of the products that were traded here in the Gulf, and probably not the most important one. Most likely, a great deal of products that were traded in the Gulf were "*invisible goods*"; products organic in nature such as textiles and foodstuffs and therefore practically invisible in the archaeological record (Potts, 1990a), it is difficult to estimate the size of the network. But I think it is safe to assume that it was on a very large scale. The archive from '*Dilmun trader'* Ea-naṣir, which was unearthed at Ur, gives us an idea on the scale of trade that was happening at this period. One text from his archives makes record of one shipment of copper from Dilmun which contained 18,33 tonnes of copper (Potts, 1990a: 224-225).

As fast as it came to overpower the Persian Gulf economy, equally fast was the demise of the Early Dilmun Society around the 18th century B.C. Its power was created by Mesopotamia's necessity for copper, but this demand changed drastically when suddenly ore from Cyprus became accessible to Babylonia (Potts, 1990a, Magee, 2014) leading to the halt of large-scale tomb construction at A'ali and the abandonment of the palace at Qala'at al-Bahrain (Højlund, 2008: 127). Though not much later after Dilmun's downfall, it again attracts the attention of the Kassite Dynasty, however, this time not as a trading partner but rather as a satellite-state at the head of which a Kassite governor is placed (Højlund, 1993, Potts, 2006). The Early Dilmun palace was in this period re-occupied and plenty Barbar-tradition pottery and Mesopotamian Kassite wares have been excavated (Højlund and Andersen, 1997: 43). A unique feature of the Middle Dilmun palace is the large amounts of bitumen that was found, all related to the construction of the building; either as the coating of walls, as bitumen-covered matting (often used for roof-construction), or simply as remnants of the fire that destroyed the building. Several of this bitumen has been sampled and analysis has shown that the samples (n=14) originate from the Hit seepages, the same type of bitumen has also been identified at Middle-Dilmun levels at Failaka (n=3) (Connan et al., 1998, Connan and Carter, 2007). And beside bitumen, a lot of Mesopotamian import materials have been identified at Failaka such as pottery, cylinder seals and cuneiform tablets (Højlund, 1987, Potts, 2010, Kjaerum, 1983, Glassner, 1984). So considering the strong Mesopotamian influence —or even dominance— over Dilmun, the presence of Mesopotamian bitumen should not come as a surprise. Beside the already mentioned sites, bitumen was also attested at al-Khor Island (Qatar). Although some pieces of bitumen were also identified in an Early Dilmun context, most of them come from Kassite- or Post Kassite contexts from which many earthenware vessels with bitumen lining have been identified (Edens, 1999, Carter and Killick, 2010). Especially the site Khor Ile-Sud seems to have been important for the production of purple-dye, and it's easy to imagine that vessels lined with bitumen –for perfect impermeability— were used for this trade. Considering the high number of Mesopotamian artefacts at this site, it is likely that the site was under Kassite control, perhaps operating from Bahrain. In this regard, the bitumen are probably also Mesopotamian in origin.

Southeast Arabian culture and society could of course not remain unchanged after its loss of direct contact with Mesopotamia and the rise of Dilmun, and consequently around 2000 B.C. the Umm an-Nar culture knows its demise. Originally, the period between 2000 and 1300 B.C. has been identified as the Wadi Suq, but C. Velde (2003) has convincingly argued that two different cultural horizons can be identified in this timespan and argues for a Wadi Suq Period (2000 B.C. – 1600 B.C.) and a Late Bronze Age (1600 B.C. – 1250 B.C.). Relatively little sites are known from this period, but it is without doubt that the Hajjar-mountains still supplied copper for Mesopotamia during the Wadi Suq period. However, the uniformity of the prior Umm an-Nar culture was lost and a decline of the collective identity is noted (Magee, 2014: 189). Even less is understood of the following due to the scarcity of (settlement) sites that have been excavated with the exceptions being Tell Abraq, al-Hamriyah and Kalba (Potts, 1991, Magee et al., 2009, Carter, 1997). For this entire period, both Wadi Suq and Late Bronze Age, bitumen has only been identified from middle 2nd millennium levels at Tell Abraq (Magee, 2014: 190). Unfortunately, the bitumen has not yet been studied in detail and no detailed information is available. The scarcity of bitumen in this period stands in contrast with the previous millennium, and it seems that the material has become less important. I have argued above that in the 3rd millennium bitumen was used to maintain and support the Mesopotamian trade networks. This changed when Dilmun starts to regulate the trade in the Gulf in the first half the 2nd millennium, followed by the eventual loss of Mesopotamian interest in Omani copper in the second half of the same millennium.

Therefore bitumen was no longer as important as before, and we see a demise in the use of this material. But undoubtedly the material still reaches southeast Arabia, as seen at Tell Abraq, but this time probably through Dilmun rather than directly from Mesopotamia. Future geochemical analyses on the Tell Abraq bitumen is a possibility and could resolve this issue. But considering the limited number of settlement sites, the possibility that we underestimate the quantities in which bitumen reaches southeast Arabia remains —although I deem it still most likely that bitumen effectively becomes more scarce in this period. Especially considering the low number of bitumen findspots in the subsequent period, which is limited to Qala'at al-Bahrain.

7.3 The Iron Age bitumen gap

In contrast to the Bronze Age when bitumen was a relatively common product, its use diminishes significantly during the Iron Age as a result of the disappearing Mesopotamian trade networks. Qala'at al-Bahrain is the only site from which bitumen has been unearthed, and the analysis of two archaeological samples dating to 700-600 B.C. indicate that this bitumen originated in Hit (Connan and Carter, 2007, Connan et al., 1998). This should not come as a complete surprise since Qala'at al-Bahrain had a privileged bond with Mesopotamia proper, and clearly maritime interactions between the two regions were maintained throughout the Iron Age.

More remarkable is the absence of bitumen from all other sites, especially because contacts between Arabian settlements and their Mesopotamian and Iranian counterparts have been identified. Assyrian cuneiform texts, for example, refer to contacts between Assyria and Dilmun and Magan, the latter two in the role of sending gifts and tributes to the former empire (Potts, 2009a: 36-37). There also seems to have been a link between southeast Arabia and Iran, evidence by the characteristic spouted vessels, columned buildings, and similarities in metal artefacts from both regions (Magee, 1998, Magee, 2002, Magee and Carter, 1999, Magee, 2005b, Magee, 2005a, Magee, 2003, Potts, 2009b). But in spite of the connections between the Persian Gulf and its neighbouring areas, bitumen was not exported to the former region. As trade between the different regions was possible, we are left to wonder why practically no bitumen is to be found dating to this period. One possibility is that there was simply no interest in the material. Another possibility relates to the nature of the integration of new overland interactions, made possible due to the domestication of the camel in the Iron Age (Magee, 2011, Magee, 2014). Bitumen and other bulk materials were easily transported using the ships of the maritime networks. Camels on the other hand were more limited to smaller, lighter cargo that could be more easily transported such luxury items. Many of the applications for which bitumen was used required large quantities of the material. It might therefore have been impractical and unprofitable to transport such amounts overland. This, combined with a possible loss of interest in the usage of bitumen, could explain the disappearance of the material in the Iron Age.

7.4 The Hellenistic trade intensifications and the re-appearance of bitumen

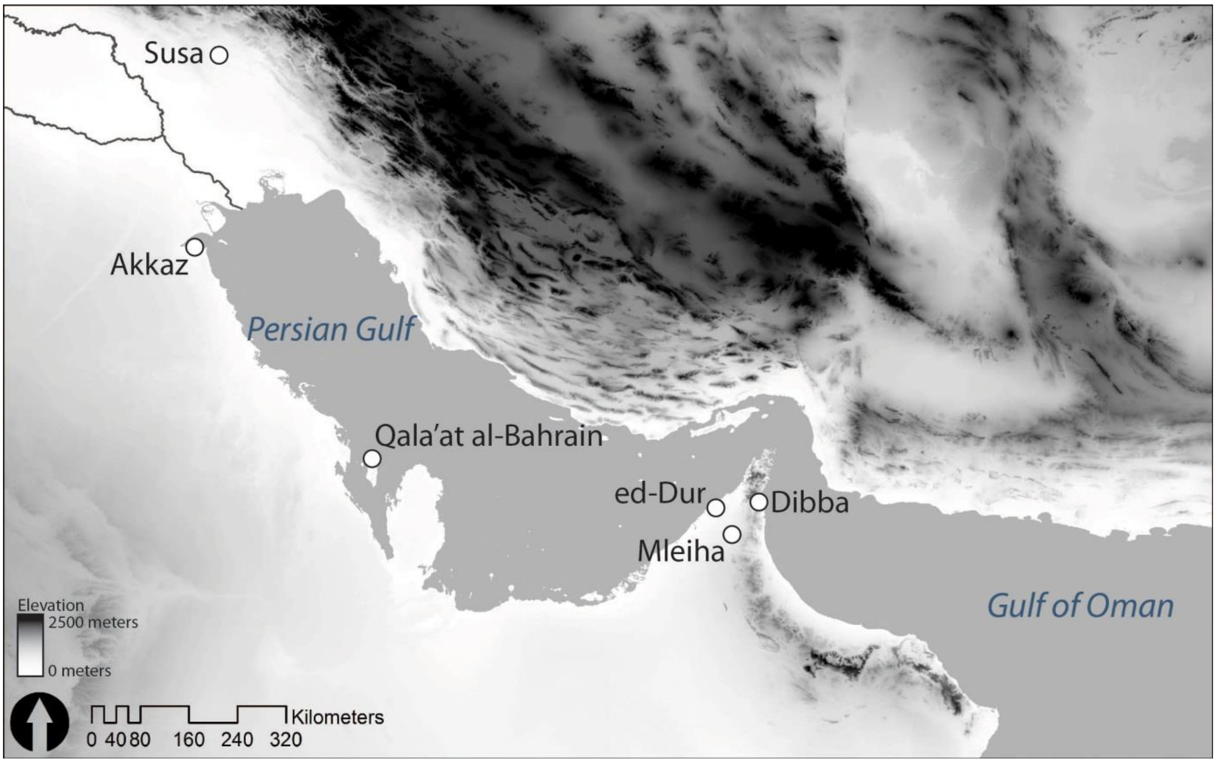

Figure 57 Hellenistic-period sites mentioned in this chapter.

There are three main observations that can be made when considering the bitumen dating to Hellenistic Periods[13]. Firstly, in contrast to previous periods, bitumen is found at all major settlements, and in considerable quantities, both in the Upper and Lower Gulf regions. Secondly, bitumen is very often (although not exclusively) found as a lining of ceramics used for transport purposes. This practice was also used in previous periods, but there is a significant increase in this type of pottery. Pottery vessels were

[13] The term Hellenistic Period is here defined as the range between the third century B.C. and the third century A.D. and not specifically linked to nor the Hellenistic- nor Parthian cultural complex.

lined with bitumen to reduce the permeability of the containers. Thus bitumen was not the actual item of trade, but rather, the *packaging*. These vessels were most likely used to transport liquids, of which wine is a viable option (Rutten, 2006). Such vessels have been attested in the Gulf at Akkaz, Failaka, Qala'at al-Bahrain, ed-Dur, Mleiha & Dibba (Rutten, 2006, Benoist et al., 2003, Connan, 2011, Jasim and Yousif, 2014, Gachet-Bizollon, 2011). They were also often used as jar-burial in Susa from the beginning of the 1st century B.C., but especially in the 1st, 2nd and third century A.D. It is likely that this would be the secondary use of these vessels and that they were primarily used as transport vessels (see Figure 58) (Boucharlat and Haerinck, 2011).

It should be noted, however, that no parallels between the Susa and Akkaz transport vessels are observable. On the other hand, these were just common transport vessels and it seems unlikely based on the available evidence that vessels of any specific shape or fabric can be attributed as being used specifically for the trade in bitumen. The third and final observation is that bitumen is found on inland sites for the first time, for example, Mleiha. These three observations can, like the changes in the Bronze Age, be explained by the changing character and nature of the economic situation in the Persian Gulf.

All of the bitumen samples analysed in this research date to first and second centuries A.D., thus from after the expansion of the Characene maritime network in the late first century B.C. (Rutten, 2006) and the Roman conquest of Egypt. This enabled the Romans to venture further eastward to India. The trade network in the Persian Gulf at this time was extensive and included areas such as southern Mesopotamia, Iran, Baluchistan and as far east as India (Seland, 2008, Hourani, 1971). This is nicely reflected in the many imported objects found at ed-Dur, one of the most extensively excavated sites dating to the first two centuries A.D (Haerinck, 2003). Salles (1993) doesn't include the Persian Gulf sphere of interaction in the vastly extensive Roman Empire network, but rather sees it as an independent, albeit often complementary, system. The incorporation of Arabia and the Persian Gulf into these networks can already be observed in the third/second century B.C. An excellent marker for this would be the finds of Rhodian amphora stamps not only in Arabia but also Susa (Monsieur et al., 2013, Monsieur et al., 2011). The situation changes in the Persian Gulf during this period, especially at an economic level. As well as the drastic increase in the scale of long-distance trade, methods also change. In particular, coins become an important method of payment. Coins are attested at many of the Pre-Islamic sites, in a lesser degree at Akkaz and Failaka (Callot, 2011), but are plentiful at ed-Dur and Mleiha (Haerinck, 1998, Potts, 1988). The discovery of a coin mould at the fortress of Mleiha (Benoist et al., 2003) also evidences the growing importance of the monetary economy.

Figure 58 Bitumen-lined transport vessels used as jar-burials in Susa.

One of the most important —and most cited— documents testifying to trade and interaction on a macro-scale is the *Periplus of the Erythraean Sea* by an unknown author in the first century A.D. This unique document mentions sewn-plank boats built in the vicinity of *Omana* and exported to Arabia (McGrail, 2001: 71). There is a general consensus that the port of Omana lies somewhere in the southern part of the Persian Gulf, although possibilities for its location have been posited of which ed-Dur (Potts, 1990b) and Dibba (Jasim, 2006) are the most recent suggestions. There is, however, no uncertainty regarding the vessels, as the Greek *Periplus* is the first document mentioning sewn-plank boats from Arabia. There is very little information available on the naval architecture of Arabia dating to this period, but sewn-plank boats are commonly known and well-documented from the Arabian Medieval period and onwards. It is possible that the introduction of this type of vessels also enabled the Arabs to more efficiently built larger ships in which more cargo could be transported. This in turn lead to an intensification of trade networks, not just with Characene, but also with Persia and India.

Changes in the pattern of bitumen trade can be understood in this context of intensifying trade networks and growing quantities of traded materials. In contrast to the Bronze Age, bitumen is no longer needed for specific purposes —such as naval architecture— or limited to areas and settlements with a privileged link to a supplier (such as Kassite Bahrain). Improved trade links and growing demand for foreign materials must have led to larger-scale exploitation of natural resources, explaining why bitumen was used at all sites, and not just the coastal settlements. The bitumen finds from ed-Dur indicate that material from multiple source areas reaches the Arabian littoral. Bitumen from Akkaz, Mleiha and Dibba seem to share the same geologic origin, an unknown seepage located somewhere in southwest Iran. It is possible that bitumen was exploited, put in transport vessels such as the ones found at Dibba, and then transported by sea via Akkaz, Bahrain, ed-Dur, finally end up in sites as far as the Gulf of Oman such as Dibba.

7.5 The transport of bitumen: raw or mixed?

We have argued in chapter 2.3.2 that bitumen mixtures were made using whatever material that was beforehand, for the mineral fraction that means that mainly ordinary sand was used as additive. But although this fraction of the mixture contains widespread and common materials, a detailed analysis may still reveal valuable information. For example, bitumen samples from Tell F6 on Failaka (French Mission) showed a high amount of carbonates in the mixture that were deliberately added and probably coming from the beaches of the island itself (Connan and Carter, 2007: 171). This indicates that the mixtures were made at their place of usage and not in Mesopotamia where the bitumen was extracted. This addresses an issue concerning the trade in bitumen; was it exported in its raw form or was it made into a mixture at the place of extraction and then transported? The data from Failaka seems to suggest the former option. In fact, (crushed) shell fragments have been identified at other sites such as Akkaz, F6 Failaka (Kuwaiti-Danish mission), al-Khidr, H3/as-Sabiyah and Dosariyah hinting to a similar practice (Connan, 2011, Connan and Carter, 2007, Van de Velde, Accepted for Publication-b, Belenová-Štolcová, 2010, Connan, 2010, Connan et al., 2005, Van de Velde et al., 2015). Unsurprisingly, all of these sites are located in a marine environment with direct access to beaches and it is no surprise that the inhabitants of these settlements used the material which was readily available.

Contrary to this, however, is the finding of large storage jars at Dibba (Sharjah Emirate, U.A.E.) in a 1-2nd century A.D. context, filled with bitumen mixtures. This is however only one isolated case, situated in a chronologically different context. It is of course possible that these things changed, that prior bitumen was traded/transported in its pure form, and later in the Hellenistic-Period it was more customary to deal in bitumen mixtures. There are just too little finds/cases to say anything conclusive on this matter.

Chapter 8 Conclusions and future perspectives

8.1 Final Conclusions

8.1.1 Major contributions to our knowledge of the bitumen trade

The initial aim of the research, presented in this book, was to track bitumen throughout the history of the Persian Gulf and use that information to identify trading networks and –contacts between the users of the bitumen (i.e. the population on several sites in the Gulf) and their supplier(s). That way, it was also possible to characterize the nature of the trade and to form several hypothesis on the specific trade in bitumen. Geochemical techniques, originally developed for the petroleum industry and the Earth Sciences, were used to detect the origin of the bitumen samples. These studies include the use of GC-MS and Stable Carbon Isotope Analysis. For my dataset, I relied upon the willingness of several researchers —currently active at in the Gulf in different parts and periods— to send their samples for analysis. I am greatly indebted to these researchers making it possible to conduct this type of research.

Every bitumen collection from a certain site was handled as an individual dataset and has generally already been published elsewhere, or is currently in publication. These different researches are bundled here in this book, with the inclusion of a dedicated introduction on the material and the specifics on the types of analysis, and a comprehensive overview of all the new research and hypotheses and ideas on both the specifics about bitumen usage per period, and on the different mechanisms of trade and the broader networks of contact it was part of.

The first dataset presented in this work is that from the 5th millennium-site of Dosariyah, located on the eastern shoreline of the Arabian Peninsula. This site is the only the second in the context of the Arabian Neolithic from which bitumen was identified and geochemically sourced. The results of the Dosariyah bitumen dataset are unique as they document the shift in the type of bitumen used; from more locally-available resources from the Burgan Hill (Kuwait) as attested at H3/as-Sabiyah, towards an interregional context that stretched as far as northern Mesopotamia. Consequently, bitumen is the second material we have undeniable proof of that was exported from Mesopotamia to the Gulf, the first being the black-on-buff pottery. This indicates that the Ubaid pottery present in the Gulf was not the only form of 'payment' or exchange good that the residents of the Arabian littoral received for their products of trade, which are presumed to be pearls.

Generally, the change in type of bitumen is to be attributed to the suppliers themselves rather than by the users of the resource. Quite remarkably, bitumen in the third millennium B.C. does not seem to have been object of trade, but rather a means of maintenance in the Mesopotamia-Magan trade. Bitumen in this context was probably limited in the naval architecture and for the upkeep of sea-going vessels. In this case, northern-Iraqi bitumen dominate the Gulf, just as they dominate in Mesopotamia at that specific period.

New analyses on new archaeological samples from Early Dilmun contexts sweeps away the old ideas on the dominance of Mesopotamian bitumen, and shows a much greater importance of Iranian bitumen, it's role apparently underestimated for a long time. How this type of bitumen entered the Persian Gulf exchange circuit is uncertain, either through southern Mesopotamia or overland in Iran to settlements such as Bushehr and then overseas. For the consequent Kassite period in the Gulf, bitumen from only 2 settlements was analysed: Tell F6 and Qala'at al-Bahrain, and bitumen from both sites coming from the seepages around Hit. This is to be expected considering the Kassite rule over the lands of Dilmun, as attested in the cuneiform lexicon.

After the disappearance of bitumen during the Iron Age in southeast Arabia, it is suddenly found at almost all Hellenistic-period sites. For the first time in the history of the Gulf, the material is now found not exclusively on coastal sites but also on the Arabian interior. This is linked to the drastic changes in the networks and economies at that time and especially the scale-up in traded materials and goods.

8.1.2 The identification of an unidentified seepage

Also bitumen from a yet unidentified seepage was identified in several samples. Whereas the bitumen from 'Akkaz was previously identified as being a mixture from material from two different sources, the exact same type of bitumen has also been attested at both Tell F6 (Bronze Age, ca. 2000 B.C.) and at Mleiha (Hellenistic period, contemporary with 'Akkaz). The appearance of bitumen with the same feature-specific chemical fingerprint at three different sites from two different periods indicates that this bitumen is not a mixture but rather an original product. As no reference data from Iranian seepages with the same specifics is identified, the location of the bitumen source remains unknown. Additionally, also other bitumen from unknown seepages has been attested at the Achaemenid sites of Tol-e Ajori and Sad-i Shahidabad. Identical samples have been found at both settlements attesting to a centralized exploitation of the natural resource.

8.2 Future perspectives

8.2.1 The obvious course to pursue

Still a gap in our knowledge on bitumen is what happens during- and after the 2nd half of the 2nd millennium B.C.. As of yet, only bitumen from Qala'at al-Bahrain has been analysed, which can hardly be called representative for the entire Gulf. A step in solving this issue would be the analysis of bitumen found in the recent joint Bryn Mawr-Tübingen University campaigns at Tell Abraq. Already, contacts have been made with the people responsible to obtain bitumen samples and have them analysed.

Also the evidence for the early periods (more specifically the Arabian Neolithic) is quite sparse as bitumen from only two sites has been identified and analysed. And although not always reported in the academic literature, bitumen is possibly identified from more Ubaid-related sites (M. Beech, pers. comm.). It is my personal thought that bitumen is a grossly underestimated material in fifth-millennium Gulf contexts and that more material will surface if more sites such as Dosariyah are excavated.

8.2.2 The socio-economic approach

Beside the logical option of pursuing to obtain more archaeological samples, the now-available datasets still offer enormous opportunities on an interpretational level. I have already briefly touched the subject of the Mesopotamian-Arabian trade, but how to put the product of bitumen in the entire Neolithic socio-economic framework? It is quite established that the black-on-buff ware in Arabia was considered as a prestigious- and luxurious good, an element of social differentiation and standing, but could we identify a similar function for the bitumen? Both products come from the same region, but can they be put on par on this level? Surely, it is difficult to see a strictly utilitarian product such as bitumen in the same role as the more elaborate pottery, but we should also consider that both products come from that *unknown and far-away land* that most of the Arabian coastline inhabitants only heard of in stories and from hearsay. In that regard, bitumen may also be seen as a product of distinction, in a context of owing something that is not indigenous but imported from far-away.

Following the same train of thought, we can continue to ponder on the role and status of bitumen during the Bronze Age. We know that many products from foreign nations such as Meluhha, Marhashi and Mesopotamia had numerous trading encounters in the Gulf exchanging rare and unique products such as precious woods, metals, lapis lazuli, etched carnelian, ivory, and many more. But assessing the role and especially the scale for what may be identified as bulk goods remains very difficult. Especially for this part of a more elaborate socio-economic study, with relations to the historic- and cuneiform records, is still in order.

Finally, the same can be said for the Hellenistic Period, where the scale of the trade is even more immense than in the Bronze Age and more reminiscent of current-day economies than ever before with the introduction of coin money and large-scale production- and transportation of goods. Again, a vital role for these studies is history and economic sciences in order to get a full understanding —if that ever is possible— of the ancient economies. Clearly, a lot is happening on that level in the Gulf. After all, the Gulf forms a crossroad for several major civilizations and different economies such as the Hellenistic Roman, Mesopotamian and even Indian.

Bibliography

AKKERMANS, P. & SCHWARTZ, G. 2003. *The Archaeology of Syria: From Complex Hunter-Gatherers to Early Urban Societies (ca. 16,000-300 BC),* Cambridge, Cambridge University Press.

ALDEN, J. R. 1982. Trade and Politics in Proto-Elamite Iran. *Current Anthropology,* 23**,** 613-640.

ALGAZE, G. 1989. The Uruk Expansion: Cross-cultural Exchange in Early Mesopotamian Civilization. *Current Anthropology,* 30**,** 571-608.

ALIZADEH, A. 2006. *The Origins of State Organizations in Prehistoric Highland Fars, Southern Iran Excavations at Tall-e Bakun,* Chicago, The University of Chicago.

ALSHARHAN, A. S. & NAIRN, A. E. M. 1997. *Sedimentary basins and petroleum geology of the Middle East,* Amsterdam ; New York, Elsevier.

ANASTASIO, S., LEBEAU, M. & SAUVAGE, M. 2004. *Atlas of Preclassical Mesopotamia,* Turnhout, Brepols.

ANDERSEN, H. H. & HØJLUND, F. 2003. *The Barbar Temples Volume 1,* Moesgaard, Jutland Archaeological Society.

ARUZ, J. 2003. *Art of the First Cities: The Third Millennium B.C. from the Mediterranean to the Indus,* New York, The MEtropolitan Museum of Art.

ASADI, A., BOUCHARLAT, R., DE SCHACHT, T., DE DAPPER, M. & UBELMANN, Y. 2009. Survey, Documentation and Ecavation on the Ancient Dams surrounding the Dasht-i Morghab Plain. A short activiteis report submitted to the Iranian center for archaeological research and the Parsa Pasargadae Research Foundation.

ASADI, A., BOUCHARLAT, R., MOHAMMADKHANI, K. & DE SCHACHT, T. 2010. Achaemeni dams to the north of Pasargadae. *Bastanpashuhi, new series,* 6**,** 135-135 (in Persian).

AZZARÀ, V.M. 2009. Domestic architecture at the Early Bronze Age sites HD-6 and RJ-2 (Ja'alan, Sultanate of Oman). *Proceedings of the Seminar for Arabian Studies*, 39, 1-15

BADEL, E. 2007. *Utilisations et circulation du bitume en Orient, durant les IVe et IIIe millénaires av. J.-C.* Master, Université Paris 1 Panthéon-Sorbonne.

BARTA, P., BENEDIKOVA, L., HAJNALOVÁ, M., MIKLIKOVÁ, Z., BELANOVA, T. & A.H., S. 2008. Al-Khidr on Failaka Island: Preliminary Results of the fieldworks at a Dilmun culture settlement in Kuwait. *Tüba-ar,* XI**,** 121-134.

BEECH, M., CUTTLER, R., MOSCROP, R., KALLWEIT, H. & MARTIN, J. 2005. New evidence for the Neolithic settlement of Marawah Island, Abu Dhabi, United Arab Emirates. *Proceedings of the Seminar for Arabian Studies,* 35**,** 37-56.

BEECH, M., ELDERS, J. & SHEPHERD, E. 2000. Reconsidering the 'Ubaid of the Southern Gulf: new results from excavations on Dalma Island, U.A.E. *Proceedings of the Seminar for Arabian Studies,* 30**,** 41-47.

BEGEMANN, F., HAUPTMANN, A., SCHMITT-STRECKER, S. & WEISGERBER, G. 2010. Lead isotope and chemical signature of copper from Oman and its occurrence in Mesopotamia and sites on the Arabian Gulf coast. *Arabian Archaeology and Epigraphy,* 21**,** 135-169.

BEGEMANN, F. & SCHMITT-STRECKER, S. 2009. Über das frühe kupfer Mesopotamians. *Iranica Antiqua,* 44**,** 1-45.

BELENOVÁ-ŠTOLCOVÁ, T. 2010. Al-Khidr, Failaka Island, State of Kuwait 2004-2008 Primary scientific report on the field research: Use of Bitumen. Unpublished Report.

BENEDIKOVA, L. & BARTA, P. 2009. A Bronze Age settlement at al-Khidr, Failakah Island, Kuwait. *Proceedings of the Seminar for Arabian Studies,* 39**,** 43-56.

BENOIST, A., MOUTON, M. & SCHIETTECATTE, J. 2003. The artefacts from the fort at Mleiha: distribution, origins, trade and dating. *Proceedings of the Seminar for Arabian Studies,* 33**,** 59-76.

BIBBY, G. 1970. *Looking for Dilmun,* London, Collins.

BLACKHAM, M. 1996. Further Investigations as to the Relationship of Samarran and Ubaid Ceramic Assemblages. *Iraq,* 58**,** 1-15.

BOEDA, E., BONILAURI, S., CONNAN, J., JARVIE, D., MERCIER, N., TOBEY, M., VALLADAS, H., AL SAKHEL, H. & MUHESEN, S. 2008. Middle Palaeolithic bitumen use at Umm el Tlel around 70 000 BP. *Antiquity,* 82**,** 853-861.

BOEDA, E., CONNAN, J., DESSORT, D., MUHESEN, S., MERCIER, N., VALLADAS, H. & TISNERAT, N. 1996. Bitumen as a hafting material on Middle Palaeolithic artefacts. *Nature,* 380**,** 336-338.

BOUCHARLAT, R., DALONGEVILLE, R., HESSE, A. & MILLET, M. 1991. Occupation humaine et environnement au 5e et au 4e millénaire sur la côte Sharjah-Umm al-Qaiwain (U.A.E.). *Arabian Archaeology and Epigraphy,* 2**,** 93-106.

BOUCHARLAT, R. & HAERINCK, E. 2011. *Tombes d'époque parthe à Susa (Chantiers de la Ville des Artisans),* Leiden, Brill.

BROWN, K. M., CONNAN, J., POSTER, N. W., VELLANOWETH, R. L., ZUMBERGE, J. & ENGEL, M. H. 2014. Sourcing archaeological asphaltum (bitumen) from the California Channel Islands to submarine seeps. *Journal of Archaeological Science,* 43**,** 66-76.

BURKHOLDER, G. 1972. Ubaid Sites and Pottery in Saudi Arabia. *Archaeology,* 25**,** 264-269.

BURKHOLDER, G. 1984. *An Arabian Collection: Artifacts from the Eastern Province,* Boulder City, GB Publications.

CALLOT, O. 2011. Chapter 7 - Akkaz (Kuwait): the Inscribed Sherd AKZ96.2441 Comments on the Characenian coins found in Kuwait (Akkaz, Umm al-Namel, Failaka). *In:* GACHET-BIZOLLON, J. (ed.) *Te Tell d'Akkaz au Koweït.* Lyon: Maison de l'Orient et de la Méditerranée.

CALVET, Y. & GACHET, J. 1990. Failaka Fouilles Françaises 1986-1988. *Travaux de la Maison de l'Orient.* Lyon: Maison de l'Orient.

CALVET, Y. & PIC, M. 1986. Un nouveau bâtiment de l'Age du Bronze sur le Tell F 6. *In:* CALVET, Y. & SALLES, J. F. (eds.) *Failaka fouilles françaises 1984-1985.* Lyon: Travauw de la masion de l'Orient.

CALVET, Y. & SALLES, J. F. 1986. Failaka Fouilles Françaises 1984-1985. *Travaux de la Maison de l'Orient.* Lyon: Maison de l'Orient.

CARTER, R. 1997. The Wadi Suq period in south-east Arabia: a reappraisal in the light of excavations at Kalba, UAE. *Proceedings of the Seminar for Arabian Studies,* 27**,** 87-98.

CARTER, R. 2003. Restructuring Bronze Age Trade: Bahrain, Southeast Arabia and the Copper Question. *In:* CRAWFORD, H. (ed.) *The Archaeology of Bahrain: The British Contribution, BAR International series 1189.* Oxford.

CARTER, R. 2005a. Chapter 6 Pottery vessels: typological analysis. *In:* KILLICK, R. & MOON, J. (eds.) *The Early Dilmun Settlement at Saar.* Ludlow: Archaeology International Ltd.

CARTER, R. 2006. Boat remains and maritime trade in the Persian Gulf during the sixth and fifth millennia BC. *Antiquity,* 80**,** 52-63.

CARTER, R. 2010. Boat-Related Finds. *In:* CARTER, R. & CRAWFORD, H. (eds.) *Maritime Interactions in the Arabian Neolithic: Evidence from H3, As-Sabiyah, an Ubaid-related site in Kuwait.* Leiden: Brill.

CARTER, R. 2014. Cultural Exchange and Identity in the Arabian-Ubaid Interaction. *Dosariyah Archaeological Research Project Workshop.* Tübingen: Unpublished.

CARTER, R. & CRAWFORD, H. 2010. *Maritime Interactions in the Arabian Neolithic: Evidence from H3, As-Sabiyah, an Ubaid-related site in Kuwait,* Leiden, Brill.

CARTER, R. & KILLICK, R. 2010. *Al-Khor Island: Investigating Coastal Exploitation in Bronze Age Qatar,* Ludlow, Moonrise Press.

CARTER, R. & PHILIP, G. 2010. Deconstructing the Ubaid. *In:* CARTER, R. & PHILIP, G. (eds.) *Beyond the Ubaid. Transformation and Integration in the Late Prehistoric Societies of the Middle East.* Chicago: The Oriental Institute of the University of Chicago.

CARTER, R. A. 2005b. The history and prehistory of pearling in the Persian Gulf. *Journal of the Economic and Social History of the Orient,* 48**,** 139-209.

CARTER, R. A. 2012. Watercraft. *In:* POTTS, D. T. (ed.) *A Companion to the Archaeology of the Ancient NEar East.* Chichester: Blackwell Publishing Ltd.

CARTER, R. A., CHALLIS, K., PRIESTMAN, S. M. N. & TOFIGHIAN, H. 2006. The Bushehr Hinterland: Results of the first season of the Iranian-British Archaeological survey of Bushehr provinde, november-december 2004 *Iran,* 44**,** 1-41.

CHARPENTIER, V., PHILLIPS, C. S. & MÉRY, S. 2012. Pearl fishing in the ancient world: 7500 BP. *Arabian Archaeology and Epigraphy,* 23**,** 1-6.

CHURLEY, M., PREST, H., ZINNIKER, D. A. & BLATT, C. Enhanced Sensitivity for Biomarker Characterization in Petroleum Using Triple Quadrupole GC/MS and Backflushing - Application Note.

CLEUZIOU, S. & MÉRY, S. 2000. In-Between the Great Powers. The Bronze Age Oman Peninsula. *In:* CLEUZIOU, S., TOSI, M. & ZARINS, J. (eds.) *Essays on the Late Prehistory of the Arabian Peninsula.* Rome: Istituto italiano per l'Africa e l'Oriente.

CLEUZIOU, S. & TOSI, M. 1994. Black Boats of Magan - Some thoughts on Bronze Age water transport in Oman and beyond from the impressed bitumen slabs of Ra's al-Junayz. *South Asian Archaeology 1993, Vols 1 and 2,* 271**,** 745-761.

CLEUZIOU, S. & TOSI, M. 2000. Ra's al-Jinz and the Prehistoric Coastal Cultures of the Ja'alan. *Journal of Oman Studies,* 11**,** 19-74.

CONNAN, J. 1988. Quelques sécrets des bitumes archéologiques de Mésopotamie revélés par les analyses de geochimie organique pétrolière. *Bulletin du Centre de recherches Elf Exploration Production,* 12.

CONNAN, J. 1999a. Study of some bituminous lumps of Mleiha - Preliminary results. *In:* MOUTON, M. (ed.) *Mleiha I - Environdment, stratégies de substistance et artisanats.* Lyon: Maison de l'Orient méditerranéen.

CONNAN, J. 1999b. Use and trade of bitumen in antiquity and prehistory: molecular archaeology reveals secrets of past civilizations. *Philosophical Transactions of the Royal Society of London Series B-Biological Sciences,* 354**,** 33-50.

CONNAN, J. 2005. La momification dans l'Egypte ancienne: le nitume et les autres ingrédients organiques des baumes de momies. *Encyclopédie religieuse de l'Univers végétal de l'Egypte ancienne.*

CONNAN, J. 2010. Appendix II: Technical Report on the Comparative Compositional and Isotopic Analysis of the H3 Bitumen. *In:* CARTER, R. & CRAWFORD, H. (eds.) *Maritime Interactions in the Arabian Neolithic: Evidence from H3, as-Sabiyah, an Ubaid-Related Site in Kuwait.* Leiden: Brill.

CONNAN, J. 2011. The bituminous mixtures of Akkaz. *In:* GACHET, J. & BIZOLLON (eds.) *Le Tell d'Akkaz au Koweit, Tell Akkaz in Kuwait.* Lyon.

CONNAN, J. 2012. *Le bitume dans l'Antiquité,* Arles, Editions Errances.

CONNAN, J. unpubl. The bitumen of ed-Dur.

CONNAN, J., BRENIQUET, C. & HUOT, J. L. 1996. Les objets bituminé de Tell el Oueili. des témoins de la diversité des résaux deéchanges commerciaux de l'Obeid 0 à l'Uruk récent. *In:* HUOT, J. L. (ed.) *Oueilo - Travaux de 1987 et 1989.* Paris: Éditions Recherche sur les Civilisations.

CONNAN, J. & CARTER, R. 2007. A geochemical study of bituminous mixtures from Failaka and Umm an-Namel (Kuwait), from the Early Dilmun to the Early Islamic period. *Arabian Archaeology and Epigraphy,* 18**,** 139-181.

CONNAN, J., CARTER, R., CRAWFORD, H., TOBEY, M., CHARRIE-DUHAUT, A., JARVIE, D., ALBRECHT, P. & NORMAN, K. 2005. A comparative geochemical study of bituminous boat remains from H3, As-Sabiyah (Kuwait), and RJ-2, Ra's al-Jinz (Oman). *Arabian Archaeology and Epigraphy,* 16**,** 21-66.

CONNAN, J. & DESCHESNE, O. 1992a. Archaeological bitumen: identification, origins and uses of an Ancient Near Eastern Material. *Materials Research Socity Symp. Proc.,* 267**,** 683-720.

CONNAN, J. & DESCHESNE, O. 1992b. Origine et altération de quelques bitumes archéologiques de Tell es-Sawwan (5500-500 avant J.-C.). *Mesopotamia,* 27**,** 47-61.

CONNAN, J. & DESCHESNE, O. 1996. *Le bitume à Suse - Collection du Musée du Lourve,* Paris et Pau, coédition Elf Aquitaine-Réunion des Musées nationaux.

CONNAN, J. & DESCHESNE, O. 2007. La bitume à Mari. *In:* MARGUERON, J.-C., ROUAULT, O. & LOMBARD, P. (eds.) *Akh Purratum 1.* Lyon: Maison de l'Orient et de la Médeterranéee.

CONNAN, J., KAVAK, O., AKIN, E., YALCIN, M. N., IMBUS, K. & ZUMBERGE, J. 2006a. Identification and origin of bitumen in Neolithic artefacts from Demirkoy Hoyuk (8100 BC): Comparison with oil seeps and crude oils from southeastern Turkey. *Organic Geochemistry,* 37**,** 1752-1767.

CONNAN, J., LOMBARD, P., KILLICK, R., HØJLUND, F., SALLES, J. F. & KHALAF, A. 1998. The archaeological bitumens of Bahrain from the Early Dilmun period (c.2200 BC) to the sixteenth century AD: a problem of sources and trade. *Arabian Archaeology and Epigraphy,* 9**,** 141-181.

CONNAN, J. & MOUTON, M. 1999. Study of Some Bituminous Lumps of Mleiha - Preliminary Results. *In:* MOUTON, M. (ed.) *Mleiha I - Environment, stratégies de subsistance et artisanats.* Lyon: Maison de l'Orient méditerranéen.

CONNAN, J., NIEUWENHUYSET, O. P., VAN AS, A. & JACOBS, L. 2004. Bitumen in early ceramic art: Bitumen-painted ceramics from Late Neolithic Tell Sabi Abyad (Syria). *Archaeometry,* 46**,** 115-124.

CONNAN, J. & NISHIAKI, Y. 2003. The Bituminous Mixtures of Tell Kosak Shamali on the Upper Euphrates (Syria) from the Early Ubaid to the Post-Ubaid: Composition of Mixtures and Origin of Bitumen. *In:* NISHIAKI, Y. & MATSUTANI, T. (eds.) *Tell Kosak Shamali: The Archaeological Investigations on the Upper Euphrates, Syria.* Oxford: Oxbow Books.

CONNAN, J. & NISSENBAUM, A. 2004. The organic geochemistry of the Hasbeya asphalt (Lebanon): comparison with asphalts from the Dead Sea area and Iraq. *Organic Geochemistry,* 35**,** 775-789.

CONNAN, J., NISSENBAUM, A. & DESSORT, D. 1992. Molecular archaeology: Export of Dead Sea asphalt to Canaan and Egypt in the Chalcolithic-Early Bronze Age (4th-3rd millennium BC). *Geochimica et Cosmochimica Acta,* 56**,** 2743-2759.

CONNAN, J., NISSENBAUM, A., IMBUS, K., ZUMBERGE, J. & MACKO, S. 2006b. Asphalt in iron age excavations from the Philistine Tel Miqne-Ekron city (Israel): Origin and trade routes. *Organic Geochemistry,* 37**,** 1768-1786.

CONNAN, J. & OATES, J. in preparation. The bitumen of Tell Brak from the Middle Uruk (c.3500 BC) to Late Bronze age (c.1280 BC): origin and trade routes. *In:* OATES, J., OATES, D., EMBERLING, G. & MCDONALD, H. (eds.) *Excavations at Tell Brak 3. The Prehistoric Periods.* Cambridge: McDonald Institure Monograph.

CONNAN, J. & OURISSON, G. 1993. De la géochimie pétrolière à l'étude des bitumes anciens : l'Archéologie Moléculaire. *Comptes Rendus des Séances de l'année 1993, Académie des Inscriptions & Belles-Lettres***,** 901-921.

CONNAN, J. & VAN DE VELDE, T. 2010. An overview of bitumen trade in the Near East from the Neolithic (c.8000 BC) to the early Islamic period. *Arabian Archaeology and Epigraphy,* 21**,** 1-19.

CONNAN, J., ZUMBERGE, J. & IMBUS, K. 2014. Bituminous mixtures of Tall-e Geser: A diversified origin of Bitumen. *In:* ALIZADEH, A. (ed.) *Ancient settlement systems and cultures in the Ram Hormuz plain, Southwestern Iran. Excavations at Tall-e Geser and Regional survey of the Ram Hormuz area.* Chicago: Oriental Institute Publications.

CONNAN, J., ZUMBERGE, J., IMBUS, K. & MOGHADDAM, A. 2008. The bituminous mixtures of Tall-e Abu Chizan: A Vth millennium BC settlement in southwestern Iran. *Organic Geochemistry,* 39**,** 1772-1789.

COUTO, D., BACQUÉ-GRAMMONT, J.-L. & TALEGHANIE, M. 2006. *Atlas Historique du Golfe Persique (XVIe-XVIIIe siècles)*, Brepols.

CRAWFORD, H. 1973. Mesopotamia's Invisible Exports in the Third Millennium B.C. *World Archaeology,* 5**,** 232-241.

CRAWFORD, H. 1998. *Dilmun and its Gulf Neighbours,* Cambridge, University Press.

CRAWFORD, H. 2005. Mesopotamia and the Gulf: The History of a Relationship. *Iraq,* 67**,** 41-46.

CRAWFORD, H. 19991. London-Bahrain Archaeological expedition: excavations at Qaar 1991. *Arabian archaeology and epigraphy,* 4**,** 1-19.

DAEMS, A. & HAERINCK, E. 2001. A burial mound at Shakhoura. *Arabian Archaeology and Epigraphy,* 12**,** 173-182.

DE MIROSCHEDJI, P. 1999. Yarmuth: The dawn of city-states in Southern Canaan. *Near Eastern Archaeology,* 62**,** 2-19.

DE SCHACHT, T., ASADI, A., UBELMANN, Y. & DE DAPPER, M. 2009. Sad-i Shahidabad 2009. Report of the second and final salvage campaign at Sad-i Shahidabad together with the research on similar monments within the Pasargadae region. Spring 2009.

DRECHSLER, P. 2011. Places of contact, spheres of interaction. The Ubaid phenomenon in the central Gulf area as seen from a first season of reinvestigations at Dosariyah (Dawsariyyah), Eastern Province, Saudi Arabia. *Proceedings of the Seminar for Arabian Studies,* 41**,** 69-82.

DRECHSLER, P. 2014. Small finds form Dosariyah. *Dosariyah Archaeological Research Project Workshop.* Tübingen: Unpublished.

EDENS, C. 1999. Khor Ile-Sud, Qatar: The Archaeology of Late Bronze Age Purple-Dye Production in the Arabian Gulf. *Iraq,* 61**,** 71-88.

EDGAR, M. C. C. 1905. Report on an excavation at Toukh el-Qaramous. *Annales du Service des Antiquités de l'Egypte,* VII**,** 205-2012.

FANTINI, A.-M. 2014. OPEC Annual Statistical Bulletin 2014. Vienna: Organization of the Petroleum Exporting Countries.

FORBES, R. J. 1958. *Studies in Early Petroleum History,* Leiden, E.J. Brill.

FORBES, R. J. 1964. *Studies in Ancient Technology, Volume I (2nd edition),* Leiden, E.J. Brill.

FRIFELT, K. 1989. 'Ubaid in the Gulf Area. *In:* HENRICKSON, E. F. & THUESEN, I. (eds.) *Upon this Foundation - The 'Ubaid reconsidered. Proceedings from the 'Ubaid symposium.* Copenhagen: Museum Tusculanum Press.

FRIFELT, K. 1995. *The Island of Umm an-Nar - Volume 2: The Third Millennium Settlement,* Moesgard, Jysk Arkaeologisk Selskab.

FUHR, B. J., HAWRELECHKO, C., HOLLOWAY, L. R. & HUANG, H. 2005. Comparison of Bitumen Fractionation Methods. *Energy & Fuels,* 19**,** 1327-1329.

GACHET-BIZOLLON, J. 2011. Chapitre V: La céramique des fouilles françaises du tell d'Akkaz (Koweït). *In:* GACHET-BIZOLLON, J. (ed.) *Le Tell d'Akkaz au Koweït.* Lyon: Maison de l'Orient et de la Méditerranée.

GLASSNER, J. J. 1984. Inscriptions cuneiforms de Failaka. *In:* SALLES, J. F. (ed.) *Failaka Fouilles Françaises 1983.* Lyon: Maison de l'Orient.

GOUAL, L. 2012. Petroleum Asphaltenes. *In:* EL-SAYED ABDUL-RAOUF, M. (ed.) *Crude Oil Emulsion- Composition Stability and Characterization.* Rijeka.

GREGG, M., BRETTELL, R. & STERN, B. 2007. Bitumen in Neolithic Iran: Biomolecular and Isotopic Evidence. *In:* GLASCOCK, M. D., SPEAKMAN, R. J. & POPELKA-FILCOFF, R. S. (eds.) *Archaeological Chemistry: Analytical Techniques and Archaeological Interpretation.* Washington: American Chemical Society.

HAERINCK, E. 1998. More pre-Islamic coins from Southeastern Arabia. *Arabian Archaeology and Epigraphy,* 9**,** 278-301.

HAERINCK, E. 2003. Internationalisation and Business in Southeast Arabia during the Late 1st century B.C./1st Century A.D.: Archaeological Evidence from Ed-Dur (Umm al-Qaiwain, U.A.E.). *In:* POTTS, D. T. (ed.) *Archaeology of the United Arab Emirates: Proceedings of the First International Conference on the Archaelogy of the UAE (Abu Dhabi).* London: Trident Press.

HARRELL, J. A. & LEWAN, M. D. 2002. Sources of mummy bitumen in ancient Egpyt and Palestine. *Archaeometry,* 44**,** 285-293.

HERMANSEN, B. 1993. Ubaid and ED pottery from five sites at 'Ain as-Sayh, Saudi Arabia. *Arabian Archaeology and Epigraphy,* 4**,** 126-144.

HØJLUND, F. 1987. *Failaka/Dilmun The second millennium settlements - Volume 2: The Bronze Age Pottery,* Moesgaard.

HØJLUND, F. 1993. The Ethnic Composition of the Population of Dilmun. *Proceedings of the Seminar for Arabian Studies,* 23**,** 1-8.

HØJLUND, F. 1995. Bitumen-coated basketry in Bahraini burial. *Arabian Archaeology and Epigraphy,* 6**,** 100-102.

HØJLUND, F. 2008. *Burial Mounds of Bahrain: Social complexity in early Dilmun,* Aarhus, Aarhus University Press.

HØJLUND, F. 2012. The Dilmun temple on Failaka, Kuwait. *Arabian Archaeology and Epigraphy,* 23**,** 152-202.

HØJLUND, F. & ABU-LABAN, A. (eds.) in publication. *Tell F6 on Failaka Island. Kuwaiti-Danish Excavations 2008-2012.,* Moesgaard: Jutland Archaeological Society.

HØJLUND, F. & ANDERSEN, H. H. 1994. *Qala'at al-Bahrain Volume 1: The Northern City Wall and the Islamic Fortress,* Moesgaard, Jutland Archaeological Society.

HØJLUND, F. & ANDERSEN, H. H. 1997. Re-occupation of the Early dilmun palace. *In:* HØJLUND, F. & ANDERSEN, H. H. (eds.) *Qala'at al-Bahrain volume 2: The Central Monumental Buildings.* Moesgaard: Jutland Archaeological Society.

HØJLUND, F. & HELLMUTH ANDERSEN, H. 1997. Qala'at al-Bahrain Volume 2 - The Central Monumental Buildings. *Jutland Archaeological Society Publucations.* Moesgaard.

HOLE, F. 1977. *Studies in the archaeological history of the Deh Luran Plain: the excavation of Chagha Sefid,* Michigan, University of Michigan.

HOLLANDER, D. & SCHWARTZ, M. 2000. Annealing, distilling, reheating and recycling : bitumen processing in the Ancient Near East. *Paleorient,* 26**,** 83-91.

HOURANI, G. F. 1971. *Arab Seafaring in the Indian Ocean in Ancient and Early Medieval Times,* New York, Princeton University Press.

HUOT, J. L. 1994. *Les premiers villageois de Mésopotamie-Du village à la ville,* Paris, Armand Colin.

JASIM, S. & YOUSIF, E. 2014. Dibba: an ancient port on the Gulf of Oman in the early Roman era. *Arabian archaeology and epigraphy,* 25**,** 50-79.

JASIM, S. A. 2006. Trade centres and commercial routes in the Arabian Gulf: Post-Hellenistic discoveries at Dibba, Sharjah, United Arab Emirates. *Arabian Archaeology and Epigraphy,* 17**,** 214-237.

KILLICK, R. & MOON, J. (eds.) 2005a. *The early Dilmun settlement at Saar,* Ludlow: Archaeology International Ltd.

KILLICK, R. & MOON, J. 2005b. Social and economic origanization. *In:* KILLICK, R. & MOON, J. (eds.) *The early Dilmun settlement at Saar.* Ludlow: Archaeology International Ltd.

KJAERUM, P. 1983. Failaka/Dilmun The Second Millennium Settlement Vol. 1 *Jutland Archaeological Society Publications.* Moesgard.

KJAERUM, P. & HØJLUND, F. 2013. *Failaka/Dilmun - The Second Millennium Settlements - Volmue 3: The Bronze Age Architecture,* Aarhus, Jutland Archaeological Society.

LAMBECK, K. 1996. Shoreline reconstructions for the Persian Gulf since the last glacial maximum. *Earth and Planetary Science Letters,* 142**,** 43-57.

LAURSEN, S. T. 2009. The decline of Magan and the rise of Dilmun: Umm an-Nar ceramics from the burial moounds of Bahrain, c. 2250-2000 BC. *Arabian archaeology and epigraphy,* 20**,** 134-155.

LAURSEN, S. T. 2011. Mesopotamian ceramics from the burial mounds of Bahrain, c.2250-1750 BC. *Arabian Archaeology and Epigraphy,* 22**,** 32-47.

LINDEMEYER, E. & MARTIN, L. 1993. *Uruk Kleinfunde III - Kleinfunde im Vorderasiatischen Museum zu Berlin: SteingefâBe und Asphalt, Farbreste, Fritte, Glas, Holz, Knochen/Elfenbein, Muschel/Perlmutt,Schnecke,* Mainz Am Rhein, Philipp Von Zabern.

LOMBARD, P. 2000. *Bahrein, la civilisation des deux mers de Dilmoun à Tylos. Exposition Institut du Monde Arabe,* Paris, Exhibitions International.

LOMBARD, P. 2004. Qal'at al-Bahreïn, mémoire d'un archipel. *In:* ÉTRANGÈRES, M. D. A. (ed.) *Archéologies, 20 ans de recherches françaises dans le mond.* Paris: Maisonneuve & Larose.

MAGEE, P. 1998. New evidence of the initial appearance of iron in southeastern Arabia. *Arabian Archaeology and Epigraphy,* 9**,** 112-117.

MAGEE, P. 2002. The Indigenous Context of Foreign Exchange between South-eastern Arabia and Iran in the Iron Age. *Journal of Oman Studies,* 12**,** 161-168.

MAGEE, P. 2003. Columned Halls, Power and Legitimisation in the Southeast Arabian Iron Age. *In:* POTTS, D., AL NABOODAH, H. & HELLYER, P. (eds.) *Archaeology of the United*

Arab Emirates: Proceedings of the First International Conference on the Archaeology of the U.A.E. London: Trident Press.

MAGEE, P. 2005a. Columned halls, bridge-spouted vessels, C 14 dates and the chronology of the East Arabian Iron Age: A response to some recent comments by O. Muscarella. *Ancient West & East,* 4**,** 160-169.

MAGEE, P. 2005b. The production, distribution and function of Iron Age Bridge-Spouted vessels in Iran and Arabia: Results from recent excavations and geochemical analysis. *Iran,* XLIII**,** 93-115.

MAGEE, P. 2011. The camel, Arabia and the role of the southern Levant in Iron Age trade - lecture given for the Departmen of Archaeology at Ghent Univeristy. Department of Archaeology - Ghent Univeristy.

MAGEE, P. 2014. *The Archaeology of Prehistoric Arabia. Adaptation and Social Formation from the Neolithic to the Iron Age,* New York, Cambridge University Press.

MAGEE, P. & CARTER, R. 1999. Agglomeration and regionalism: Southeastern Arabia between 1400 and 1100 BC. *Arabian Archaeology and Epigraphy,* 10**,** 161-179.

MAGEE, P., UERPMANN, H. P., UERPMANN, M., JASIM, S. A., HANDEL, M., BARBER, D., FRITZ, C. & HAMMER, E. 2009. Multi-disciplinary research on the past human ecology of the east Arabian coast: excavations at Hamriya and Tell Abraq (Emirate of Sharjah, United Arab Emirates). *Arabian Archaeology and Epigraphy,* 20**,** 18-29.

MAISELS, C. K. 1993. *The Near East: Archaeology in the 'Cradle of Civilization',* London, Routledge.

MARCUCCI, L. G., GENCHI, F., BADEL, E. & TOSI, M. 2011. Recent investigations at the prehistoric site RH-5 (Ra's al-Hamra, Muscat, Sultanate of Oman). *Proceedings of the Seminar for Arabian Studies,* 41**,** 201-222.

MARSCHNER, R.-F., DUFFY, L. J. & WRIGHT, H. 1978. Asphalts from Ancient town sites in Southwestern Iran. *Paleorient,* 4**,** 97-112.

MASRY, A. H. 1974. *Prehistory in northeastern Arabia: the problem of interregional interaction,* Miami, Field research projects.

MASRY, A. H. 1997. *Prehistory in Northeastern Arabia: The Problem of Interregional Interaction,* London, Routledge.

MATTHEWS, R. 2001. From Kuwait to Ras al-Khaimah: Ubaid connections in the Gulf. *In:* BRENIQUET, C. & KEPINSKI, C. (eds.) *Études Mésopotamiennes. Recueil de texted offert à Jean-Louis Huot.* Paris.

MAURER, J., MÖHRING, T. & RULLKÖTTERL, J. 2002. Plant lipids and Fossil Hydrocarbons in Embalming Material of Roman Period Mummies from the Dakhleh Oasis, Western Desert, Egypt. *Journal of Archaeological Science,* 29**,** 751-762.

MCCLURE, H. A. & AL-SHAIKH, N. A. 1993. Palaeogeography of an 'Ubaid archaeological site, Saudi Arabia. *Arabian Archaeology and Epigraphy,* 4**,** 107-125.

MCCOWN, D. E., HAINES, R. C. & BIGGS, R. C. 1978. Nippur II, The north temple and sounding E. Chicago.

MCGRAIL, S. 2001. *Boats of the World: From the Stone Age to Medieval Times,* Oxford, Oxford University Press.

MENGHIN, O. & AMER, M. (eds.) 1936. *The Excavations of the Egyptian university in the Neolithic site at Maadi, Second Preliminary Report, Season 1932,* Caïro: Gov.Press.

MERY, S. & CHARPENTIER, V. 2013. Neolithic material cultures of Oman and the Gulf seashores from 5500-44500 BCE. *Arabian Archaeology and Epigraphy,* 24**,** 73-78.

MÉRY, S., CHARPENTIER, V., AUXIETTE, G. & PELLE, E. 2009. A dugong bone mound: the Neolithic ritual stite on Akab in Umm al-Quwain, United Arab Emirates. *Antiquity,* 83**,** 696-708.

MONSIEUR, P., BOUCHARLAT, R. & HAERINCK, E. 2011. Amphores Grecques Timbrées découvertes à Suse (SO-Iran). *Iranica Antiqua,* XLVI**,** 161-192.

MONSIEUR, P., OVERLAET, B., JASIM, S. A., YOUSIF, E. & HAERINCK, E. 2013. Rhodian amphora stamps found in Mleiha (Sharjah, UAE): old and recent finds. *Arabian Archaeology and Epigraphy,* 24**,** 208-223.

MOON, J. 2005. Tools, weapons, utensils and ornaments. *In:* KILLICK, R. & MOON, J. (eds.) *The early Dilmun settlement at Saar.* Ludlow: Archaeology International Ltd.

MOOREY, P. R. S. 1994. Ancient Mesopotamian Materials and Industries. The Archaeological Evidence. Oxford: Clarendon Press.

NISSENBAUM, A. 1978. Dead Sea asphalt-Historical aspects. *American Association of Petroleum Geologists Bulletin,* 62**,** 837-844.

NISSENBAUM, A. 1994. Utilization of Dead Sea asphalt through history. *Rev.Chem.Engng,* 9**,** 365-383.

NISSENBAUM, A. & BUCKLEY, S. 2012. Dead Sea Asphalt in Ancient Egyptian Mummies - Why? *archaeometry,* 55**,** 563-568.

NISSENBAUM, A. & GOLDBERG, M. 1980. Asphalts, heavy oils, ozocerite and gases from the Dead Sea Basin. *Organic Chemistry,* 2**,** 167-180.

NISSENBAUM, A., SERBAN, A., AMIRAN, R. & ILAN, O. 1984. Dead Sea Asphalt from the Excavations in Tel Arad and Small Tell Malhata. *Paléorient,* 10**,** 157-161.

OATES, J. 1969. Choga Mami, 1967-68: A Preliminary Report. *Iraq,* 31.

OATES, J. 1993. Trade and Power in the Fifth and Fourth Millennia B.C.: New Evidence from Northern Mesopotamia. *World Archaeology,* 24**,** 403-423.

OATES, J., DAVIDSON, T. E., KAMILLI, D. & MCKERRELL, H. 1977. Seafaring merchants of Ur? *Antiquity,* 51**,** 221-234.

OATES, J. & OATES, D. 2004. The role of Exchange Relations in the Origins of Mesopotamian Civilization. *In:* CHERRY, J. F., SCARRE, C. & SHENNAN, S. (eds.) *Explaining Social Change: Studies in Honour of Coling Renfrew.* Cambridge: McDonald Institute for Archaeological Research.

OCHSENSCHLAGER, E. L. 1992. Ethnographic Evidence for wood, boats, bitumen and reeds in Southern Iraq, Ethnoarchaeology at al-Hiba. *In:* POSTGATE, N. & POWELL, M. A. (eds.) *Trees and Timber in Mesopotomia.* Sumerian Agriculture Group.

OLIJDAM, E. 2014. From rags to riches: three crucial steps in DIlmun's rise fo fame. *Proceedings of the Seminar for Arabian Studies,* 44**,** 277-286.

OPPENHEIM, A. L. 1954. The Seafaring Merchants of Ur. *Journal of the American Orienta Society,* 74**,** 6-17.

PETERS, K. E., WALTERS, C. C. & MOLDOWAN, J. M. 2005a. *The Biomarker Guide Volume 1: Biomarkers and Isotopes in the Environment and Human History,* New York, Cambridge University Press.

PETERS, K. E., WALTERS, C. C. & MOLDOWAN, J. M. 2005b. *The Biomarker Guide: Volume 2, Biomarkers and Isotopes in Petroleum Systems and Earth History,* New York, Cambridge University Press.

PETRIE, C. A., CHAVERDI, A. A. & SEYEDIN, M. 2005. From anshan to Dilmun and Magan: The spatial and temporal distribution of Kaftari and Katari-related Ceramic Vessels. *Iran,* XLIII**,** 49-86.

PÉZARD, M. 1914. *Mission à Bender-Bouchir Documents archéologiques et èpigraphiques,* Paris.

PHILLIPS, C. 2002. Prehistoric Middens and a Cemetery from the Southern Arabian Gulf. *In:* CLEUZIOU, S., TOSI, M. & ZARINS, J. (eds.) *Essays on the Late Prehistory of the Arabian Peninsula.* Rome: Instituto Italiano per l'Africa e l'Oriente.

PIC, M. 2008. La céramique du tell F6, fouilles françaises de 1984 à 1988. *In:* CALVET, Y. & PIC, M. (eds.) *Failaka Fouilles Françaises 1984-1988: Matériel céramique du temple-tour et épigraphie.* Lyon: Maison de l'Orient et de la Méditerranée.

PINCÉ, P., OVERLAET, B., HAERINCK E. Unpubl. Report. Belgian Archaeological Research in Mleiha (Sharjah, U.A.E.): Field season 2009-2012.

POTTS, D. T. 1988. Arabia and the Kingdom of Characene. *In:* POTTS, D. T. (ed.) *Araby the Blest: Studies in Arabian Archaeology.* Copenhagen: the Carsten Niebuhr Institute of Ancient Near Eastern Studies, University of Copenhagen Museum.

POTTS, D. T. 1990a. *The Arabian Gulf in Antiquity Volume I - From Prehistory to the Fall of the Achaemenid Empire,* Oxford, Clarendon Press.

POTTS, D. T. 1990b. *The Arabian Gulf in Antiquity Volume II - From Alexander the Great to the Coming of Islam,* Oxford, Clarendon Press.

POTTS, D. T. 1991. *Further Excavations at Tell Abraq. The 1990 Season,* Copenhagen, Munksgaard.

POTTS, D. T. 1993a. The Late Prehistoric, Protohistoric, and Early Historic Periods in Eastern Arabia (ca. 5000-1200 B.C.). *Journal of World Prehistory,* 7**,** 163-212.

POTTS, D. T. 1993b. Rethinking some aspects of trade in the Arabian Gulf. *World Archaeology,* 24**,** 423-440.

POTTS, D. T. 1995. Watercraft of the Lower Sea. *In:* FINKBEINER, U., DITTMANN, R. & HAUPTMANN, H. (eds.) *Beiträge zur Kulturgeschichte Vorderasiens. Festschrift für Rainer Michael Boehmer.* Mainz: von Zabern.

POTTS, D. T. 1997. *Mesopotamian Civilization : The Material Foundations,* New York, Cornell University Press.

POTTS, D. T. 1999. *The Archaeology of Elam: Formation and Transformation of an Ancient Iranian State*, Cambridge University Press.

POTTS, D. T. 2000. *Ancient Magan. The Secrets of Tell Abraq,* London, Trident.

POTTS, D. T. 2006. Elamites and Kassistes in the Persian Gulf. *Journal of Near Eastern Studies***,** 110-119.

POTTS, D. T. 2009a. The Archaeology and Early History of the Persian Gulf. *In:* POTTER, L. G. (ed.) *The Persian Gulf in History.* New York: Palgrave Macmillan.

POTTS, D. T. 2009b. Urartian and Assyrian echoes at Saruq al-Hadid (Emirate of Dubai). *Liwa,* 1, 3-9.

POTTS, D. T. 2010. Cylinder seals and their use in the Arabian Peninsula. *Arabian Archaeology and Epigraphy,* 18, 20-40.

POTTS, T. F. 1993c. Patterns of trade in third-millennium BC Mesopotamia and Iran. *World Archaeology,* 24, 379-402.

POURNELLE, J. 2003. *Marshland of Cities: Deltaic Landscapes and the Evolution of Early Mesopotamian Civilization.* Doctor of Philisophy, University of California.

RATNAGER, S. 1981. *Encounters: the Westerly Trade of the Harappa Civilization,* Delhi.

RATNAGER, S. 2004. *Trading Encounters, From the Euphrates to the Indus in the Bronze Age,* Oxford, Oxford Univeristy Press.

RENIERS, G. 2008. *Fundamentele begrippen van de organische chemie,* Leuven, Acco.

RIVA, J. P. 2013. *Petroleum* [Online]. Encyclopaedia Britannica. Available: http://www.britannica.com/EBchecked/topic/454269/petroleum [Accessed 27/03/2015.

ROAF, M. 1974. Excavation at Al Markh, Bahrain : A fish midden of the fourth millennium B.C. *Paléorient,* 2, 499-501.

ROAF, M. 1976. Excavations at Al Markh, Bahrain. *Proceedings of the Seminar for Arabian Studies,* 6, 144-160.

ROAF, M. & GALBRAITH, J. 1994. Pottery and p-values: "Seafaring merchants of Ur?" re-examined. *Antiquity,* 68, 770-783.

ROUX, G. 1992. *Ancient Iraq; third edition,* London, Penguin Group.

ROWLAND, I. D. & NOBLE HOWE, T. 1999. *Vitruvius: Ten Books on Architecture,* New York, Cambridge University Press.

RULLKÖTTER, J. & NISSENBAUM, A. 1988. Dead Sea asphalt in Egyptian mummies: molecular evidence. *Naturwissenschaften,* 75, 618-621.

RULLKOTTER, J., SPIRO, B. & NISSENBAUM, A. 1985. Biological markers characteristics of oils and asphalts from carbonate source rocks in a rapidly subsiding Graben, Dead-Sea, Israel. *Geochimica Et Cosmochimica Acta,* 49, 1357-1370.

RUTTEN, K. 2006. *Het aardewerk van ed-Dur (Umm al-Qaiwain, V.A.E.) uit de late 1ste eeuw v. tot de vroege 2de eeuw n. Chr. Technologische, typologische en vergelijkende studie met een analyse van de ruimtelijke verspreiding en handel in en voorbij de Perzische Golf.* PhD, Universiteit Gent.

SALLES, J. F. 1984. Failaka Fouilles Françaises 1983. *Travaux de la Maison de l'Orient.* Lyon: Maison de l'Orient.

SALLES, J. F. 1993. The Periplus of the Erythraean Sea and the Arab-Persian Gulf. *Topoi,* 3, 493-523.

SCHWARTZ, M. & HOLLANDER, D. 2008. Bulk stable carbon and deuterium isotope analyses of bitumen artefacts from Hacinebi Tepe, Turkey: reconstructing broad economic patterns of the Uruk expansion. *Journal of Archaeological Science,* 35, 3144-3158.

SELAND, E. H. 2008. The Indian ships at Moscha and the Indo-Arabian trading circuit. *Proceedings of the Seminar for Arabian Studies,* 38, 283-287.

SPIRO, B., WELTE, D. H., RULLKÖTTER, J. & SCHAEFER, R. G. 1983. Asphalts, oils and bituminous rocks from the Dead Sea area - a geochemical correlation study. *American Association of Petroleum Geologists Bulletin,* 67**,** 1163-1175.

STEIN, G. J. 1994. Economy, Ritual and Power in 'Ubaid Mesopotamia. *In:* STEIN, G. J. & ROTHMAN, M. S. (eds.) *Chiefdoms and Early States in the Near East: The Organizational Dynamics of Complexity.* Madison: Prehistory Press.

STEIN, G. J. 2010. Local identities and interaction spheres: Modeling regional variation in the Ubaid horizon. *In:* CARTER, R. & PHILIP, G. (eds.) *Beyond the Ubaid.* Chicago: The Oriental Institute of the University of Chicago.

STEIN, G. J., HOLLANDER, D. & SCHWARTZ, M. 1999. Reconstructiong Mesopotamian Exchange Networks in the 4th Millennium BC: Geochemical and Archaeological Analyses of Bitumen Artifacts from Hacinebi Tepe, Turkey. *Paléorient,* 25**,** 67-82.

STEIN, G. J. & ÖZBAL, R. 2007. A Tale of Two oikumenai: Variation in the Expansionary Dynamics of 'Ubaid and Uruk Mesopotamia. *In:* STONE, E. C. (ed.) *Settlement and Society: Essays Dedicated to Robert McCormick Adams.* Chicago: The Oriental Institute of the University of Chicago.

STEINKELLER, P. 2004. Towards a Definition of Private Economic Activity in Third Millennium Babylonia. *In:* ROLLINGER, R. & ULF, C. (eds.) *Commerce and Monetary Systems in the Ancient World: Means of Transmission and Cultural Interaction.* Munich: Franz Steiner Verlag.

STEINKELLER, P. 2006. New Light on Marhaši and its contacts with Makkan and Babylonia. *Journal of Magan Studies,* 1**,** 1-17.

STERN, B., CONNAN, J., BLAKELOCK, E., JACKMAN, R., CONINGHAM, R. A. E. & HERON, C. 2008. From Susa to Anuradhapura: Reconstructing aspects of trade and exchange in bitumen-coated ceramic vessels between Iran and Sri lanka from the third to the ninth centuries AD. *Archaeometry,* 50**,** 409-428.

STOL, M. 2012. Bitumen in Ancient Mesopotamia. The Textual Evidence. *Bibliotheca Orientalis,* 69**,** 48-60.

TENGBERG, M. & POTTS, D. T. 1999. (gis)mes.ma-ga-na (Dalbergia sissoo Roxb.) at Tell-Abraq. *Arabian Archaeology and Epigraphy,* 10**,** 129-133.

THE ETCSL PROJECT. 2006. *The building of Ningirsu's temple (Gudea, cylinders A and B)* [Online]. The Oxford University. Available: http://etcsl.orinst.ox.ac.uk/cgi-bin/etcsl.cgi?text=t.2.1.7# [Accessed 06/04/2015 2015].

THESIGER, W. 1964. *The Marsh Arabs,* London, Longmans, Green and co ltd.

UERPMANN, M. 2003. The Dark Millennium - Remarks on the Final Stone Age in the Emirates and Oman. *In:* POTTS, D. T., AL NABOODAH, H. & HELLYER, P. (eds.) *Archaeology of the United Arab Emirates - Proceedings of the First International Conference on the Archaeology of the U.A.E.* Abu Dhabi: Trident Press.

UERPMANN, M. & UERPMANN, H. P. 1996. 'Ubaid pottery in the Eastern Gulf - new evidence from Umm al-Qawain (U.A.E.). *Arabian Archaeology and Epigraphy,* 7**,** 125-139.

VAN DE VELDE, T. 2015. Digging into the Ubaid-Period bitumen from Dosariyah. *Proceedings of the Seminar for Arabian Studies,* 45.

VAN DE VELDE, T. Accepted for Publication-a. Geochemical analysis on the bitumen lining of a vessel excavated from Tumulus B-5 by Peter B. Cornwall. *In:* PORTER, B. W. & BOUTIN, A. T. (eds.) *Embodying Ancient Dilmun: The Peter B. Cornwall Expedition to Bahrain and Saudi Arabia.* Boston: American School of Oriental Research Archaeological Report Series.

VAN DE VELDE, T. Accepted for Publication-b. Sourcing the bitumen from Tell F6. *In:* HØJLUND, F. & ABU-LABAN, A. (eds.) *Tell F6 on Failaka Island. Kuwaiti-Danish Excavations 2008-2012.* Moesgaard: Jutland Archaeological Society.

VAN DE VELDE, T. & BODÉ, S. Accepted for Publication. Appendix 2: Analysis of Bitumen from the Royal Mounds. *In:* LAURSEN, S. T. (ed.) *The Rise of Kingship and the Early Dilmun State in Bahrain.* Moesgaard: Jutland Archaeological Society.

VAN DE VELDE, T., DE VRIEZE, M., SURMONT, P., BODÉ, S. & DRECHSLER, P. 2015. A geochemical study on the bitumen from Dosariyah (Saudi-Arabia): tracking Neolithic-period bitumen in the Persian Gulf. *submitted for the Journal of Archaeological Science*.

VAN DE VELDE, T., DE VRIEZE, M., SURMONT, P., BODÉ, S. & DRECHSLER, P. 2015. A geochemical study on the bitumen from Dosariyah (Saudi-Arabia): tracking Neolithic-period bitumen in the Persian Gulf. *Journal of Archaeological Science,* 57**,** 248-256.

VELDE, C. 2003. Wadi Suq and Late Bronze Age in the Oman Peninsula. *In:* POTTS, D., AL NABOODAH, H. & HELLYER, P. (eds.) *Archaeology of the United Arab Emirates - Proceedings of the First International Conference on the Archaeology of the U.A.E.* Abu dhabi: Trident Press.

VOGT, B. 1994. In Search for Coastal Sites in Prehistoric Makkan: mid-Holocene 'shell eaters' in the coastal desert of Ras al-Khaimah, U.A.E. *In:* KENOYER, J. M. (ed.) *From Sumer to Meluhha: Contributions to the Archaeology of South and West Asia in Memory of George F. Dales, Jr.* Wisconsin.

VOSMER, T. 2000. Model of a Third Millennium BC Reed Boat based on evidence from Ra's al-Jinz. *Journal of Near Eastern Studies,* 11**,** 149-153.

VOSMER, T. The Naval Architecture of Early Bronze Age Reed-built Boats of the Arabian Sea. *In:* POTTS, D., AL NABOODAH, H. & HELLYER, P., eds. Proceedings of the First International Conference on the Archaeology of the U.A.E., 2001 Abu Dhabi. Trident Press, 151-157.

WALLACE, D., HENRY, D., PONGAR, K. & ZIMMERMAN, D. 1987. Evaluation of some open column chromatographic methods for separation of bitumen components. *Fuel,* 66**,** 44-50.

WANG, Z. Chemical Fingerprinting of Spilled or Discharged Petroleum - Methods and Factors Affecting Petroleum Fingerprints in the Environment. The IV Workshop of the Cooperative Network on Recovery of Contaminated Areas by Petroleum Activities - RECUPETRO, 2008 Salvador, Brazil.

WANG, Z. & FINGAS, M. 2005. Oil and Petroleum Product Fingerprinting Analysis by Gas Chromatographic Techniques. *In:* NOLLET, L. M. L. (ed.) *Chromatographic Analysis of the Environment, Third Edition.* CRC Press.

WARBURTON, D. A. 2011. What might the Bronze Age world-System Look Like? *In:* WILKINSON, T. C., SHERRATT, S. & BENNET, J. (eds.) *Interweaving worlds. Systemic Interactions in Eurasia, 7th to 1st Millennia BC.* Oxford: Oxbow Books.

WEEKS, L. 1999. Lead isotope analyses from Tell Abraq, United Arab Emirates: New data regarding the 'tin problem' in Western Asia. *Antiquity,* 73**,** 49-64.

WEEKS, L. 2003. *Early Metallurgy of the Persian Gulf, Technology, Trade and the Bronze Age World,* Leiden, Brill Academic Publishers.

WEEKS, L. & COLLERSON, K. 2005. Archaeometallurgical Studies. *In:* KILLICK, R. & MOON, J. (eds.) *The Early Dilmun Settlement at Saar.* Ludlow: Archaeology International Ltd.

WEEKS, L. R. 1997. Prehistoric Metallurgy at Tell Abraq, U.A.E. *Arabian Archaeology and Epigraphy,* 8**,** 11-85.

WOOLLEY, S. L. 1955. *Ur Excavations Volume IV - The Early Periods,* Philidalphia, Allen, Lane & Scott.

WRIGHT, H. & BERGER, E. 1981. *An early town on the Deh Luran Plain: excavations at Tepe Farukhabad*, Ann Arbor: Museum of Antrhopology, University of Michigan.

WRIGHT, H. & JOHNSON, G. A. 1975. Population, Exchange and early state formaton in Southwesteren Iran. *American Anthropologist New Series,* 77**,** 267-289.

ZARINS, J. 2008. Magan Shipbuilders at the Ur III Lagash State Dockyards (2062-2025 B.C.). *In:* OLIJDAM, E. & SPOOR, R. (eds.) *Intercultural Relations between South and Southwest Asia.* BAR S1826 ed. Oxford: Archaepress.

Appendix I: Achaemenid bitumen

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Introduction

Five bitumen samples were selected for geochemical analyses in order to determine their origin from the archaeological site of Tol-e Ajori. These will be discussed in this chapter together with two samples from Sad-i Shahidabad. This chapter will outline the analyses and give a detailed report and interpretation of the results. Two types of analyses were conducted on the five bitumen samples from Tol-e Ajori; investigation of molecular ratios on the Saturated Hydrocarbon fraction obtained through GC-MS analysis, followed by stable carbon isotope analyses on the Asphaltene fraction.

The bitumen from Tol-e Ajori

Bitumen was found in large numbers all over the site, especially present in collapse/destruction layers. The usage of bitumen on the site seems to have focused mainly as a building mortar for the construction of the monumental building. Five bitumen pieces were chosen for sampling and send to Ghent for geochemical analyses. Three samples came from destruction layers, one from a course of brick, and one from the inside of a jar (see Table 9).

Table 9 The five bitumen samples used for geochemical screening.

Trench	SU	Sample ID	Bitumen Sample	Comments
1	SU072	0067	BS1	
1	SU06	0068	BS2	Bitumen sample from inside of the jar
2	SU009	0069	BS3	
2	SU14	0070	BS4	
1	SU41	0071	BS5	Mortar of brick course, sampled while cleaning

Analysis of the samples

The sample preparation as outline in chapter 3.4, and the measured values are shown in Table 10. Prior to this research, no bitumen from Achaemenid contexts was ever published, but it was suspected that the bitumen used at Tol-e Ajori came from southwest Iran, an area holding a lot of seepages from which plenty of reference material is published. Analyses of $\delta^{13}C$ also limited the potential seepages from which this bitumen may have come from. Numerous settlements have been supplied by one- or more seepages in southwestern Iran, and bitumen from several of them have been published in great detail and create excellent references for this research. The bitumen used as reference in this work came from Abu Chizan (Connan et al., 2008), Tall-e Geser (Connan et al., 2014), Umm an-Namel (Connan and Carter, 2007), Ali Kosh, Chageh Sefid & Tepe Tula'I (Gregg et al., 2007).

Beside Tol-e Ajori, Sad-i Shahidabad is the only site from which bitumen has been studied in detail with sourcing the material as primary aim. Shahidabad is located in the Fars-province of Iran and has been identified as a dam and water-related control structure, erected during the Early Achaemenid period (corresponding to the reigns of Cyrus, Cambyses and Darius) (Asadi et al., 2009, De Schacht et al., 2009, Asadi et al., 2010). Bitumen was attested all over in the joints of the limestone construction blocks to waterproof the entire structure. Two bitumen samples from this site has been analysed in Ghent and we will refer to these samples as well in this report.

Table 10 Measured values of the Tol-e Ajori bitumen samples.

Sample	$\delta^{13}C$	Ts/Tm	GCRN/C_{30}	GCRN/31R
BS1	-28,6	0,21	0,19	0,51
BS2	-28,41	0,72	0,27	0,45
BS3	-27,8	0,11	0,19	0,59
BS4	-28,85	0,08	0,19	0,57
BS5	-28,63	0,12	0,19	0,6

The origin of the bitumen samples

None of the five samples show any traces of 18α(H)-oleanane, a chemical compound occurring mainly in Late Cretaceous or younger source rock, and for the geographical area here at hand more specifically from the Padbeh source rock formation (Peters et al., 2005b: 572, Connan, 2012: 117). Figure 20 shows the known Iranian bitumen seepages and their oleanane/ C_{30} ratios.

Other determining molecular ratios for fingerprinting archaeological bitumen are 18α-22,29,30-trisnorneohopane to 17α-22,29,30-trisnorhopane (Ts/Tm), Gammacerane to 17α,21β-hopane (GCRN/C_{30}αβ-Hopane), and Gammacerane to C_{31}22R hopane (GCRN/31R).

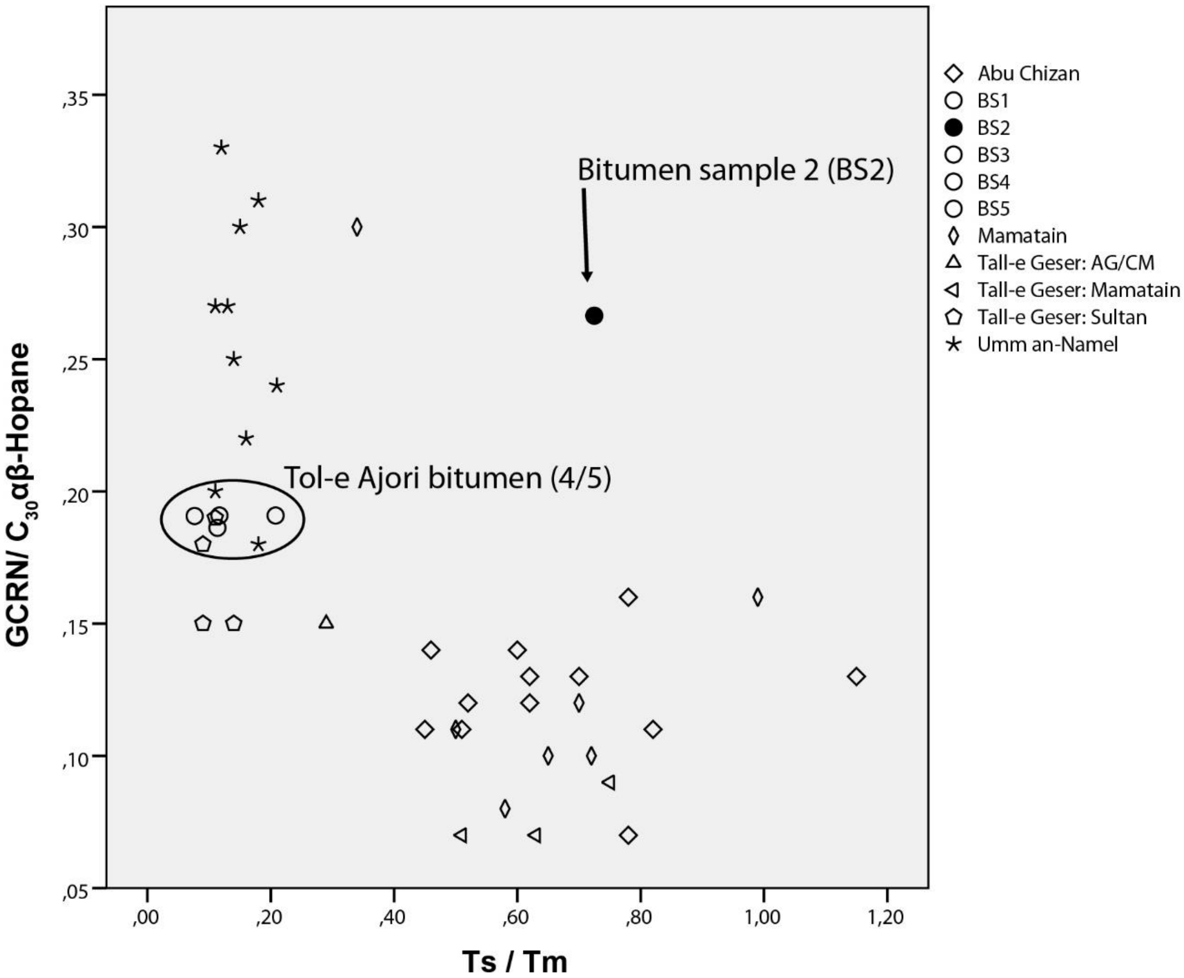

Figure 59 Ts/Tm to Gammacerane/C_{30}-Hopane scatter plot. The samples from Tall-e Geser are relabeled to their respective source areas.

Figure 59 shows the plotted Ts/Tm to GCRN/C_{30} ratios, and we can see that four of the samples show a strong similarity to the Hit bitumen (Iraq) from Umm an-Namel (Kuwait), and the bitumen excavated at Tall-e Geser from the Sultan seepage (Iran, Luristan province). All other bitumen from Iranian sites and seepages tend to have higher Ts/Tm- and lower GCRN/C_{30} ratios, or show an obvious presence of 18α(H)-oleanane. The molecular ratios and the measured $\delta^{13}C$ of the Tol-e Ajori samples are also similar to those from measurements performed on a bitumen samples from Sad-i Shahidabad, and bitumen from both sites seems to have come from the same seepage.

There is however one obvious outlier in this dataset (Bitumen Sample 2, also marked differently in the legends of the scatter plots) which doesn't seem to have a relation with any other sample. Based upon measurements on five samples, Tol-e Ajori seems to be supplied by bitumen from at least 2 different seepages.

A scatter plot of measured $\delta^{13}C$ values to the Ts/Tm ratio can be seen in Figure 60 and shows additional info to the previous scatter plot. The Iranian bitumen seepages with their $\delta^{13}C$ values are shown in Figure 18. Three of the bitumen samples show a remarkable lower $\delta^{13}C$ value than any other bitumen from the archaeological sites (or even from any natural seepage for that matter, see Figure 60). Statistical analyses conducted on the dataset (hierarchical clustering and k-means clustering) doesn't favour a direct correlation of the Tol-e Ajori samples with either the Hit-bitumen from Umm an-Namel, nor with the archaeological samples coming from the Sultan seepage. It is therefore most likely that the bitumen used here come from another, yet unidentified seepage.

We should always bear in mind that archaeological bitumen to oil seep correlation are limited by the set of references for which geochemical data is available (Connan, 2011), and that seepages active during Antiquity are not necessarily still providing resources. Unfortunately there is no detailed information available whether on (active) seepages in the Persepolis-area in Antiquity, and a systematic survey of this area for this sole purpose is yet to be achieved. That is more than likely also the reason why there are absolutely no reference samples with the same molecular ratios as sample BS2, the only exception being one of the two archaeological samples from Sad-i Shahidabad. Quite remarkably; this sample from Shahidabad also shows a clear presence of oleanane, which is not the case for sample BS2 from Tol-e Ajori. Either this sample from Shahidabad is coming from yet another source with very similar molecular ratios as BS2 from Tol-e Ajori, or bitumen from another seepage —with oleanane present— was added to the bitumen mixture used at Shahidabad.

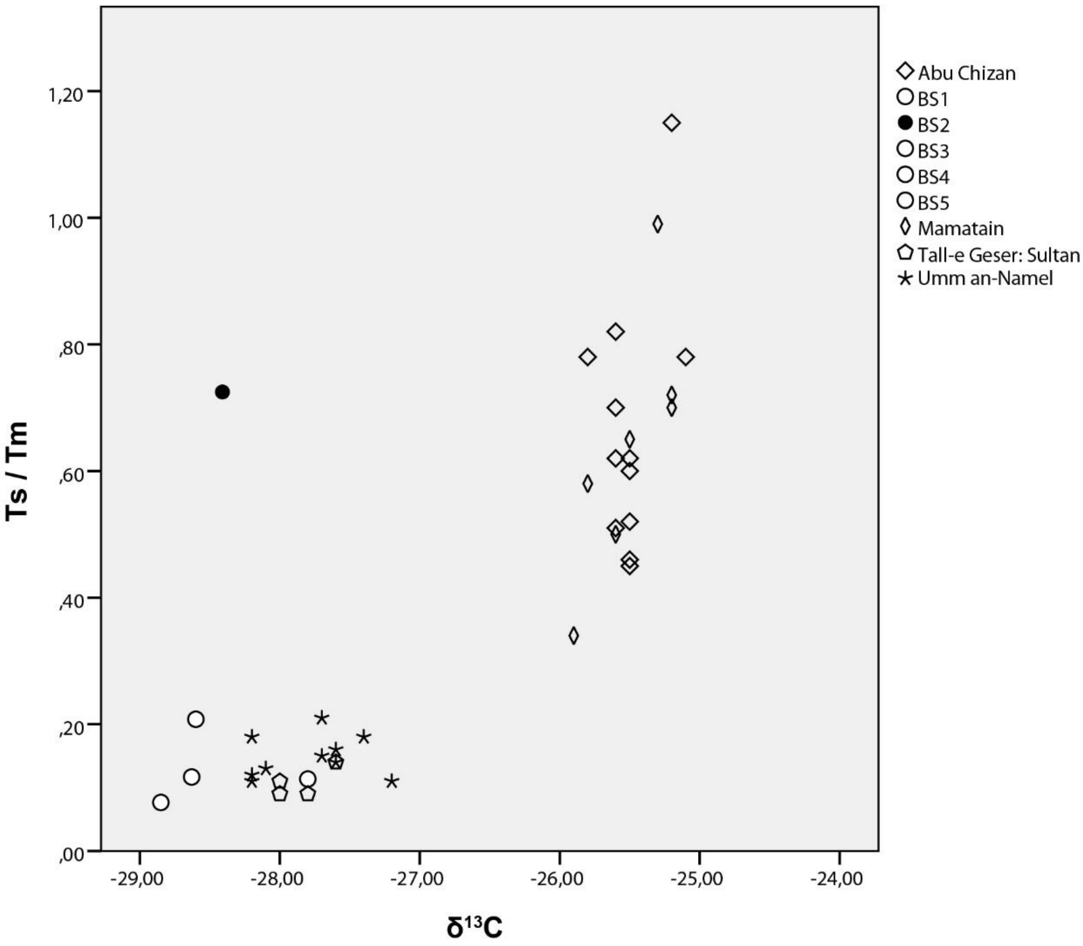

Figure 60 Scatter plot of $\delta^{13}C$ (expressed in ‰) to Ts/Tm. Besides the obvious outlier BS2, it also appears that the other samples fall without the range of most other reference samples.

Conclusions

No positive match was found for the bitumen used at Tol-e Ajori with any of the reference seepages, nor with any sourced bitumen from any other archaeological site. We should bear in mind though we do not own reference data from all active bitumen seepages in Antiquity. The analyses conducted on the bitumen samples from Tol-e Ajori did however point out that the site was supplied with bitumen from at least two different seepages, and that both the molecular ratios and the $\delta^{13}C$ values are in accordance with bitumen retrieved from the contemporary site of Sad-i Shahidabad. The main usage of bitumen both at Tol-e Ajori and Sad-i Shahidabad was as a constructing material; either as a mortar or as a seam seal in joints. Both applications require large quantities of bitumen and therefore a steady flow of natural resources. In such case, a local source without the need of transport over large distance would be best suited. That would also explain why the bitumen doesn’t match with any of the seepages on- and around the Deh Luran- and Susiana plain, nor with those from the famous bitumen sources at Hit in present-day Iraq, both of which located a substantial distance from both Tol-e Ajori and Sad-i Shahidabad.

Appendix II: Bitumen in the 3rd millennium of the Near East

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Introduction

Bitumen is nowadays known as a by-product of the refining of crude oil, and used for road construction and roofing. It is formed by the thermal degradation of organic material deep beneath the surface and accumulates in underground reservoirs. These reservoirs can release their bitumen through cracks and faults, allowing the bitumen to find a natural way to the earth's surface. Where bitumen crops out on the surface, it can easily be gathered.

Petroleum occurred naturally in various locations in the Ancient Near East, and people living in these places started exporting the bitumen quite early. It was seldom used in its natural form and other materials were added to create a mixture which was easier to work with.

Usage

The adhesive and waterproof properties of bitumen proved useful in many applications. It was used as glue for numerous everyday purposes (gluing flint in sickles, mending broken pottery, ...) as well as for elaborate pieces of art (see Figure 61). In architecture, it was used as a mortar between brickwork, but also as a waterproof protection for monuments, rooms, wells, cisterns, quays, etc. In fact, bitumen was used for all kind of purposes for which waterproofing was necessary; such as coating of earthenware vessels, storage pits, and baskets (see Figure 62). A special usage for bitumen is reserved in the construction of ships, where the material was used to waterproof and strengthen the hull.

Figure 61 The Standard of Ur, where bitumen was used to glue shell and lapis lazuli on a wood core (image ©Trustees of the British Museum).

Figure 62 Remains of bitumen-covered basket from Mleiha (1^{st}-2^{nd} century A.D., Emirate of Sharjah, UAE), the imprint of the vegetal core is still visible. Photo taken by the author, with thanks to B. Overlaet and E. Haerinck of the Belgian Archaeological Mission at Mleiha.

Bitumen trade

For archaeologists, a main advantage of bitumen is that this material can often be sourced to the original place of extraction by means of chemical analysis . If bitumen is found on an archaeological site, and if its source is identified, we can reconstruct trade networks and patterns of interaction.

The most important occurrences of bitumen during antiquity are highlighted on the map. These seepages were known and exploited by man. Bitumen could surface at various locations (i.e. pools, puddles), but if it was formed in the same underground reservoirs, these various sources share the same chemical footprint. Archaeological bitumen samples are analysed according to geochemical techniques of petroleum geochemistry, and their results compared to reference samples in order to establish correlations. Many results of these studies will trace the samples back to one of the major areas.

Bitumen prior to the Early Bronze Age

Working with bitumen is a very simple process. The sites of Umm el Tlel and Hummal prove that the material was already exploited in the Middle Palaeolithic Period (ca. 70,000 BP). Bitumen became more widely in use in the Near East during the 7th millennium B.C.E. Already during this period, bitumen was used at sites far away from the natural sources. From the Ubaid 3 period onward (mid 6th millennium), we have evidence for organized trade in bitumen originating from the Mosul area. Also, this period provides us with the earliest evidence of boats caulked with bitumen, this to make stronger and more waterproof.

The Uruk period seems to have been an important period for the bitumen business. Not only was the material more used in monumental architecture (Moorey, 1994: 335), it also seems to have originated from the Hit area alongside the River Euphrates, rather than from the Mosul seepages. Bitumen is found at settlements alongside this river, more than anywhere else in northern Mesopotamia (with the exception of Tell Brak, located near the Khabur river). Sites such as Hacinebi Tepe, Djebel Aruda, Sheikh Hassan, and Habuba Kabira were supplied with bitumen from the Hit area. The site of Tell Brak seems to have been an exception and was supplied with bitumen from both the Hit and the Mosul area.

Perhaps not incidentally, copper from Anatolia travelled southwards to Mesopotamia (Begemann and Schmitt-Strecker, 2009: 23) via this river, and it has been suggested that the same trading posts/networks were used for both products (and likely a wider array of resources and products) (Connan and Van de Velde, 2010).

Third millennium bitumen

Unfortunately, not many bitumen samples from third millennium Mesopotamian contexts have been analysed. But when looking at northern Mesopotamian settlements where bitumen has been found, we see that in the Jemdet Nasr period these sites are located mainly alongside the Balikh River rather than the Euphrates. The data for this interpretation has been derived from (Anastasio et al., 2004). Could this imply, again, a change in bitumen-supplier for Mesopotamia? Bitumen from the Mosul area has been found at various 5th-, 4th- and 3rd millennium settlements located on the shorelines of the Persian Gulf (such as Umm an-Nar & Ra's al-Jinz), implying that bitumen from this source was extracted on a large scale and widely used. Cuneiform texts from the Gudea and Ur III periods also hint to this area as the main bitumen-supplier for Mesopotamia (Stol, 2012: 56). So far, the evidence seems to hint to Mosul as the main bitumen source during this period.

However, all the bitumen from Mari (an entire sequence from the Early Dynastic I period till the early 2nd millennium) was sourced to the nearby Hit area (Connan and Deschesne, 2007). Surprisingly, the bitumen from early 2nd millennium Qala'at al-Bahrain and Tall Sa'ad (more commonly known as site F6, Failaka) also show a clear Hit signature, in contrast to other sites in the Persian Gulf (Connan and Carter, 2007), indicating direct contact between Dilmun and Mari.

Second millennium and younger texts (cuneiform, classic authors) refer to Hit as the most important bitumen-bearing area. Hammurabi points out to Zimri-Lim "*[...]Exactly because of the [bitumen] I want this city*" (Stol, 2012: 57). Remarkably, bituminous mortar starts appearing at Babylon for the first time during the reign of Hammurabi (Moorey, 1994: 72), and it might be a safe bet to assume that this bitumen originated from the Hit area. This bitumen kept on being used at Babylon, as Neo-Babylonian period samples indicate (Connan, 1988).

Not only in Mesopotamia, but also in southwest Iran was bitumen a popular product.. Intensive interaction between the Deh Luran valley and the Susiana plain is known from the Proto-Elamite period (3200 – 2700 B.C.E.) onwards, which is also reflected in the material evidence. Bitumen from the Susiana plain has been discovered at settlements in the Deh Luran, and vice versa (Marschner et al., 1978: 110). Also, samples from Susa and Tall-e Geser evidence the import of bitumen from different sources (Connan, 2012, Connan et al., 2014), hinting at an extensive organization of raw materials. Iranian bitumen has not been attested in 3rd and 2nd millennium Mesopotamia settlements, but was found at the Early Dilmun settlement at Saar (Bahrain) (Connan et al., 1998).

The Eastern Mediterranean

Although bitumen from the Dead Sea has so far never been attested in Mesopotamia, Iran, or the Persian Gulf, it has been found at several Bronze Age sites in the Eastern Mediterranean. It seems like this bitumen had its own specific trade network, focusing on the area south & west of the Dead Sea and Egypt (Connan et al., 1992). Recent studies also point to Hasbeya oil seeps as a possible supplier of the bitumen used in this area (Connan and Nissenbaum, 2004).

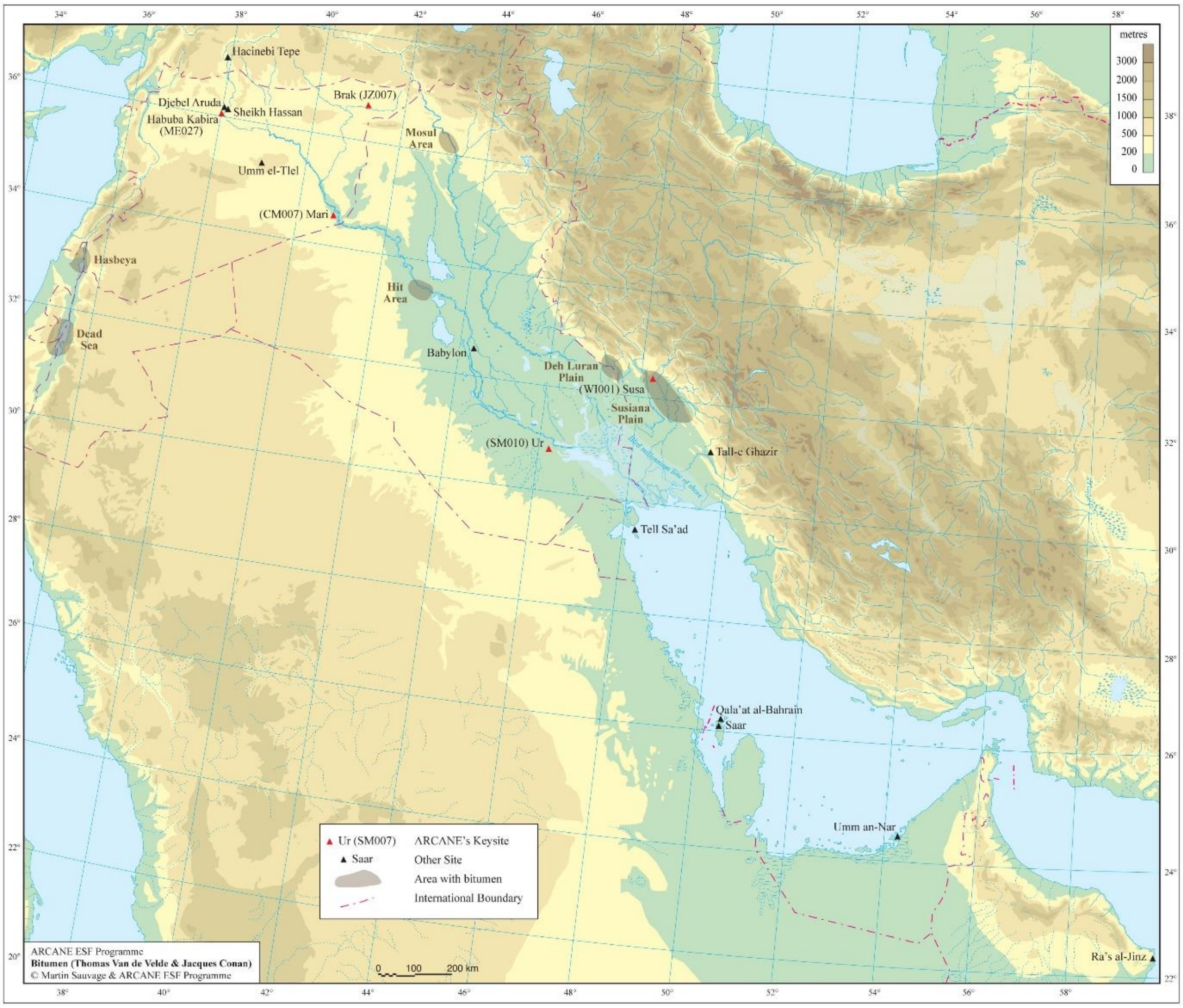

Figure 63 All sites mentioned in this chapter. ©Martin Sauvage & Arcane ESF Programma.